# 「Introduction of A New Principle in the Theory of Magnetism III」の報告に当って

東大理　飯田修一

本論文は物性研究に既に報告した同一主題の論文，Ⅰ[1)]，Ⅱ[2)]，に引き続くものであって，物性研究誌上で話題になった[3),4)]ものである。その際公表の近いことを約束したにも関らず今日に至ったのは，当初Ⅰ,Ⅱの基礎となる重要論文として日本物理学会の Journal に公表を予定し，1975年5月投稿されたが，レフェリー過程に非常な時間が掛り，現時点でも yes が出ない状況に直面したので，物性研究の読者諸子の御批判を得て早急に Journal 公表を得るよう方針を変更したことによる。従って，討議に値するものとしての Journal 発表に支障すると考えられる箇所を発見された場合は早急に飯田宛コメントされるよう希望させて戴く。

なほ本論文はすべて古典物理学の手法で記述されているが，古典物理学の手法は，量子物理学の手法と矛盾するものではなく，完全に平行するものであって，巨視的事象に関する限り，古典物理学の手法は正しい。とくに本論文は $\mathbf{r}$，$\mathbf{v}$，即ち $q_r$，$\dot{q}_r$ を使用するラグランジアンの手法で記述されるが，$L(q_r, \dot{q}_r)$ が与えられたとき

$$p_r = \frac{\partial L}{\partial \dot{q}_r} \quad , \quad \frac{d}{dt}\left(\frac{\partial L}{\partial \dot{q}_r}\right) - \frac{\partial L}{\partial q_r} = 0 \tag{1}$$

のラグランジュの方程式は $q_r$，$\dot{q}_r$，$p_r$ を演算子と考えることにより量子論的にも完全に厳密に正しく[5)]，そのことは Schrödinger 表示でも Heisenberg 表示でも関係ない。量子力学の体系として加えるべき唯一の条件は正準交換関係

$$[q_r, \ p_s] = i\hbar\, \delta_{rs} \tag{2}$$

だけである。この証明は必ずしも既存の文献で十分と言い難いように思えたので，筆者として独自に厳密に証明していることを附記させて戴く。勿論ハミルトニアンが使える場合

$$\frac{dq_r}{dt} = \frac{\partial H}{\partial p_r} \quad , \quad \frac{dp_r}{dt} = -\frac{\partial H}{\partial q_r} \tag{3}$$

も演算子間の関係式として表示に関係なく厳密に成り立つのである。

なほ Part Ⅰ[1)]，Ⅱ[2)] には誤植等が多いのですが，Ⅱ には本質的なミスが一ケ所あり，先の文献[1)]と本論文中でその訂正を行って居ります。御注意下さい。又 Joumal レフェリーとの間には 60回に亙るメモの往復があり提起された疑問点は既に精密に解析されていますが，紙面の関係でそれらのすべてを掲載することは出来なかったことを附記させて戴きます。

## 文　　献

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# Introduction of A New Principle in the Theory of Magnetism, III

## Fundamental Meaning of The Vector Potential, The Magnetic Energy, The Hamiltonian and The Meissner Effect

**Shuichi IIDA**

Department of Physics
University of Tokyo
Tokyo, JAPAN

## Synopsis

Fundamental meaning of the vector potential $\mathbf{A}(\mathbf{r})$ and presence of a new energy term for an electron, $-ev\cdot\mathbf{A}(\mathbf{r})/c$, are clarified for a classical electron gas. An essential advantage of the Lagrangian treatment over the Hamiltonian treatment for the thermostatistics of a macroscopically inhomogeneous magnetic system is emphasized. A new general principle in irreversible thermodynamics called the transient energy principle has been introduced. We conclude that Miss van Leeuwen's theorem is wrong, the Meissner effect is a classical property of a perfect conductor, and new separate thermodynamics should be used for the paramagnet, the diamagnet, and the superconductor.

## § 1. Introduction

In recent several years, the author has made an extensive study for reorganizing the classical electromagnetism in terms of the Maxwell-Lorentz electromagnetism and have reached to several conclusions that are different from the present common understandings. The results are being published in "Bussei Kenkyu" in English as a series of papers, on the introduction of a new principle in the theory of magnetism.[1),2)] This paper is the presentation of part III of the series,[3)] which is most important and written more carefully.

We believe that, in previous thermostatistical theories of magnetism, there has been an insufficient understanding for the magnetic interaction energy of the externally applied magnetic field. The physical meaning of the vector potential A has been also not well understood. We have concluded that Miss van Leeuwen's theorem on the absence of diamagnetism in classical systems[4),5),6)] is wrong and a classical perfect conductor, if once created quantum mechanically, should show the Meissner effect automatically[1),2)].

For the purpose of simplification all the description in this paper will be made in terms of classical physics. However, we believe that, regarding most of the equations as operator equations, the essential physical results derived must be duely effective in quantum physics.

The description will be made in MKS rationalized Gauss unit system,[7),8)] for convenience*.

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*We call this system MKSP system in which P stands for physical[8)].

§ 2. Relativistic Derivation of the Orbit-Orbit Mutual Interaction Energy of Moving Electrons

Suppose we have a single charge, q, at rest, the electromagnetic potential four vector** is

$$\{a_0(r_0),\ i\phi_0(r_0)\} = \{0,\ i\frac{q}{4\pi r_0}\} \tag{1}$$

This four vector transforms into

$$\{a(r), i\phi(r)\} = \{\frac{qv}{4\pi cr\sqrt{1-\frac{(v\times \hat{r})^2}{c^2}}},\ i\frac{q}{4\pi r\sqrt{1-\frac{(v\times \hat{r})^2}{c^2}}}\} \tag{2}$$

in frame K, in which frame $K_0$ is moving with the velocity v. Here $\hat{\mathbf{r}}$ is the unit vector along **r** in K, and these expressions are known as the Lienard-Wiechert potentials for an uniform motion.[9] Introducing the nonuniform part of the motion and using an approximation, we get

$$\{a(r), i\phi(r)\} = \{\frac{qv}{4\pi cr},\ i\frac{q}{4\pi r}[1+\frac{(v\times \hat{r})^2 - \dot{v}\cdot r}{2c^2}]\} \tag{3}$$

in which $\dot{v}$ is the acceleration vector of the charge. Up to this approximation, the retarded and advanced potentials coincide exactly, showing that no emission nor absorption of free electromagnetic radiation is involved[10),11)]. The degree of the approximation of Eq. (5), however, becomes worse when r becomes large.

Now the Coulomb gauge expression of Eq. (3) is

$$\{a^C(r), i\phi^C(r)\} = \{\frac{qv}{4\pi cr} + \frac{q\hat{r}\times(\hat{r}\times v)}{8\pi cr},\ i\frac{q}{4\pi r}\} \tag{4}$$

and the acceleration vector $\dot{v}$ disappears.[12)]

When we are interested in a system which contains many moving charges which are almost in thermally stationary state, the contribution from one of these charges to the macroscopic Lorentz electromagnetic potentials at a long distance is most adequately represented by Eq. (4). Because, in the assumed situation, the retarded and advanced ambiguity[13),14)] is not present and, the second term of $a^C$ in Eq. (4)[15),16)] becomes statistically ineffective at long distances.

(Proof is easy.)

When there are two moving electrons in a free space, the total electromagnetic energy of the system[10),12),17),18),19),20)] is

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***i* is used as the unit imaginary number for the four space.

$$\iiint_\infty \frac{e^2+h^2}{2}dV = \iiint_\infty \frac{(e_1+e_2)^2+(h_1+h_2)^2}{2}dV$$

$$= \iiint_\infty \frac{e_1^2+h_1^2}{2}dV + \iiint_\infty \frac{e_2^2+h_2^2}{2}dV + \iiint_\infty e_1\cdot e_2\, dV + \iiint_\infty h_1\cdot h_2\, dV$$

$$= \frac{mc^2}{\sqrt{1-(\frac{v_1}{c})^2}} + \frac{mc^2}{\sqrt{1-(\frac{v_2}{c})^2}} + \frac{e^2}{4\pi r_{12}} + \frac{e^2 v_1\cdot v_2}{4\pi r_{12}c^2} + \frac{e^2(v_1 \times \hat{r}_{12})\cdot(v_2 \times \hat{r}_{21})}{8\pi c^2 r_{12}} \tag{5}$$

Here $\hat{r}_{ij}$ is the unit vector along $r_{ij}$, and we have concluded that the kinetic energy of an electron amalgamates the electromagnetic self-energy of the electron.

In these calculations, for the sake of simplification we have disregarded the spin-spin and spin-orbit magnetic interactions completely.

## § 3. Derivation of the Macroscopic Vector Potential and the Associated Magnetic Energy of A System

Let us extend our calculation into a total system, in which there are many electrons and nuclei in a quasi-stationary state. It is to be noted that this system includes both specimens and the source of the applied magnetic field. Then the electromagnetic energy, $U_{E.M.}$ is

$$U_{E.M.} = \iiint_\infty \frac{e^2+h^2}{2}dV = \iiint_\infty \frac{(\sum_i e_i)^2+(\sum_i h_i)^2}{2}dV$$

$$= \sum_i \iiint_\infty \frac{e_i^2+h_i^2}{2}dV + \sum_{i\neq j}\iiint_\infty \frac{e_i\cdot e_j}{2}dV + \sum_{i\neq j}\iiint \frac{h_i\cdot h_j}{2}dV \tag{6}$$

Applying the introduced amalgamation procedure, we get as the total energy U,

$$U = \sum_i m_i c^2 + \sum_i \frac{m_i v_i^2}{2} + \sum_i \frac{3m_i v_i^4}{8c^2} + \sum_{i\neq j}\frac{q_i q_j}{8\pi r_{ij}} + \sum_{i\neq j}\frac{q_i q_j}{8\pi r_{ij}}\frac{v_i\cdot v_j}{c^2}$$

$$- \frac{1}{2c^2}\iiint\left(\sum_{i\neq j}\frac{\partial\phi_i}{\partial t}\cdot\frac{\partial\phi_j}{\partial t}\right)dV \tag{7}$$

Let us define the Maxwell-Lorentz electromagnetic potentials in the Coulomb and Lorentz gauges as[7])

$$\phi^C(r_\lambda) = \sum_i \phi_i^C(r_\lambda) \tag{8}$$

$$a(r_\lambda) = \sum_i a_i(r_\lambda) \tag{9}$$

Then, the Maxwell macroscopic electromagnetic potentials in the Lorentz gauge can be represented as

$$\varphi(r_\lambda) = \frac{1}{\Delta V_\lambda} \iiint_{\Delta V_\lambda} \phi^C(r)\, dV \tag{10}$$

$$A(r_\lambda) = \frac{1}{\Delta V_\lambda} \iiint_{\Delta V_\lambda} a(r)\, dV \tag{11}$$

Here, $\Delta V_\lambda$ is the volume at the location, $r_\lambda$, which is small macroscopically but large microscopically. Then

$$\phi^C(r) = \varphi(r) + \phi'(r) \tag{12}$$

$$a(r) = A(r) + a'(r) \tag{13}$$

in which

$$\left.\begin{aligned} \bar{\bar{\phi}}'(r) &= 0 \\ \bar{\bar{a}}'(r) &= 0 \end{aligned}\right\} \tag{14}$$

Here, double bar means the volume average on $\Delta V$. With these notations, we have

$$\begin{aligned} \sum_{i \neq j} \frac{q_i q_j}{8\pi r_{ij}} &= \frac{1}{2} \sum_i q_i \sum_{i \neq j}^{j} \frac{q_j}{4\pi r_{ij}} = \frac{1}{2} \sum_i q_i [\phi^C(r_i) - \phi_i^C(r_i)] \\ &= \frac{1}{2} \sum_i q_i [\varphi(r_i) + \phi'(r_i) - \phi_i^C(r_i)] \\ &= \iiint \frac{\rho(r)\varphi(r)}{2}\, dV + \frac{1}{2} \sum_i q_i [\phi'(r_i) - \phi_i^C(r_i)] \end{aligned} \tag{15}$$

The last term of Eq. (15) represents the electrostatic part of the formation energy, $U_{E.S.f.}$, for the constituent atoms and the lattice. Of course $\phi'(r_i)$ and $\phi_i^C(r_i)$ are almost identical at the locations near $r_i$[7] and only the second order term is effective. Then we get

$$\sum_{i \neq j} \frac{q_i q_j}{8\pi r_{ij}} = \iiint_\infty \frac{\rho(\mathbf{r})\varphi(\mathbf{r})}{2} dV + U_{E.S.f.} \tag{16}$$

Similarly,

$$\sum_{i \neq j} \frac{q_i q_j}{8\pi r_{ij}} \frac{\mathbf{v}_i \cdot \mathbf{v}_j}{c^2} = \frac{1}{2} \sum_i q_i \frac{\mathbf{v}_i}{c} \cdot \mathbf{A}(\mathbf{r}_i) + \frac{1}{2} \sum_i q_i \frac{\mathbf{v}_i}{c} [\mathbf{a}'(\mathbf{r}_i) - \mathbf{a}_i(\mathbf{r}_i)]$$

$$= \iiint \frac{\mathbf{j}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r})}{2c} dV + U_{M.f.} \tag{17}$$

$$-\frac{1}{2c^2} \iiint \left( \sum_{i \neq j} \frac{\partial \phi_i}{\partial t} \frac{\partial \phi_j}{\partial t} \right) dV = -\frac{1}{2c^2} \iiint \left( \frac{\partial \varphi}{\partial t} \right)^2 dV + U_{E.D.f.}$$

$$= \left[ \sum_{i > j} \frac{q_i q_j}{8\pi r_{ij} c^2} (\mathbf{v}_i \times \hat{\mathbf{r}}_{ij}) \cdot (\mathbf{v}_j \times \hat{\mathbf{r}}_{ji}) \right] \tag{18}$$

Here, $U_{M.f.}$ represents the magnetic part of the formation energy, and $U_{E.D.f.}$ is the short range electrodynamic part of the formation energy.

In this way, we get the total energy of the system,[21),22)]

$$U = \sum_i m_i c^2 + \sum_i \frac{m_i v_i^2}{2} + \sum_i \frac{3 m_i v_i^4}{8c^2} + \iiint_\infty \frac{\rho(\mathbf{r})\varphi(\mathbf{r})}{2} dV + \iiint_\infty \frac{\mathbf{j}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r})}{2c} dV$$

$$-\frac{1}{2c^2} \iiint \left( \frac{\partial \varphi}{\partial t} \right)^2 dV + U_{E.S.f.} + U_{M.f.} + U_{E.D.f.} \tag{19}$$

and a part of the energy which is dependent on a single electron,

$$mc^2 + \frac{m v_i^2}{2} + \frac{3 m v_i^4}{8c^2} - e\varphi(\mathbf{r}_i) - \frac{e\mathbf{v}_i \cdot \mathbf{A}(\mathbf{r}_i)}{c} \qquad (e > 0)$$

$$-e[\phi'(\mathbf{r}_i) - \phi_i^C(\mathbf{r}_i)] - \frac{e\mathbf{v}_i \cdot [\mathbf{a}'(\mathbf{r}_i) - \mathbf{a}_i(\mathbf{r}_i)]}{c}$$

$$-\frac{e\mathbf{v}_i}{c} \cdot \sum_{j \neq i} \frac{-[q_j \mathbf{v}_j + (q_j \mathbf{v}_j \cdot \hat{\mathbf{r}}_{ji}) \hat{\mathbf{r}}_{ij}]}{8\pi r_{ij} c} \tag{20}$$

When the considered electron is a conduction electron, the usual first order approximation is to replace the short range interaction terms with time independent ripple potentials having the periodicity of the atomic arrangement. Then the effective expression of Eq. (20) is

$$U = \frac{mv_i^2}{2} - e\varphi^*(\mathbf{r}_i) - \frac{e\mathbf{v}_i \cdot \mathbf{A}^*(\mathbf{r}_i)}{c} \qquad (21)$$

in which the last term is not included in the usual conventional Hamiltonian, but it can have a value of several tens of electron volts at the surface area of a specimen.

It is noted that in general there is no large freedom for the Lorentz covariant macroscopic electromagnetic potentials $\{\mathbf{A}(\mathbf{r}, t), i\varphi(\mathbf{r}, t)\}$ in a given physical system. The only mathematical freedom is due to $\psi(\mathbf{r}, t)$[15),16)] such as

$$\mathbf{A}' = \mathbf{A} - \nabla\psi, \qquad \varphi' = \varphi + \frac{1}{c}\frac{\partial\psi}{\partial t}, \qquad \Delta\psi - \frac{1}{c^2}\frac{\partial^2\psi}{\partial t^2} = 0 \ . \qquad (22)$$

However, $\psi(\mathbf{r}, t)$ can be represented by the values of $\psi(\mathbf{r}, t)$ at the very remote surface S of a large volume V which includes the entire system as [14)]

$$\psi(\mathbf{r}_1, t_1) = \iint_S \left[\psi(\mathbf{r}_2, t_2)\frac{\nabla_2 r_{12}}{4\pi r_{12}^2} + \frac{\nabla_2\psi(\mathbf{r}_2, t_2)}{4\pi r_{12}} + \frac{\nabla_2 r_{12}}{4\pi r_{12}}\frac{\partial\psi(\mathbf{r}_2 t_2)}{c\partial t_2}\right]_{t_2 = t_1 - \frac{r_{12}}{c}} \cdot d\mathbf{S}_2 \qquad (23)$$

$$= \iint_S \left[\psi(\mathbf{r}_2, t_2)\frac{\nabla_2 r_{12}}{4\pi r_{12}^2} + \frac{\nabla_2\psi(\mathbf{r}_2, t_2)}{4\pi r_{12}} - \frac{\nabla_2 r_{12}}{4\pi r_{12}}\frac{\partial\psi(\mathbf{r}_2 t_2)^2}{c\partial t_2}\right]_{t_2 = t_1 + \frac{r_{12}}{c}} \cdot d\mathbf{S}_2 \qquad (24)$$

and it represents either a ghost wave packet which comes from outside and escapes to infinity, or stationary $\psi(\mathbf{r})$ and $\nabla\psi(\mathbf{r})$ both of which become $\pm\infty$ at remote places[21),22)]. It is evident that these $\psi(\mathbf{r}, t)$ cannot have the symmetry of the system considered in this paper and should be put as zero physically.

## § 4. Energy Transfer Relation Associated with the Motion of An Electron in A Magnetic field

When an electron is in a constant magnetic field $\mathbf{H}$, the force on the electron is determined by a local property of $\mathbf{A}$, being characterized by $\mathbf{H} = \nabla \times \mathbf{A}$, but, the energy of Eq. (21) must experience continuous change, the amount of which must be cancelled by the corresponding reversed change of the energy in a certain part of the system.

For further analysis, however, we have to differentiate whether the time for the electromagnetic information to travel between the electron and the source of the magnetic field is shorter or longer than the time for which the state of the electron, as seen from the source, can be assumed unchanged.

The distance $\Delta L$, by which the electromagnetic information can travel in the $\Delta\theta$ radian of the orbital motion of a cyclotron motion, can be calculated as

$$\Delta L = \frac{mc^2}{eH_0}\Delta\theta = 2 \times 10^{-(0 \sim 3)}\Delta\theta \text{ m} = 20 \text{ cm} \sim 0.2 \text{ mm} \tag{25}$$

for the magnetic field and $\Delta\theta$ of

$$H_0 = 10^{1 \sim 4} \text{ Oe}, \quad \Delta\theta = 0.1 \text{ radian}. \tag{26}$$

Let us consider first the case in which

$$\Delta L > L_M \tag{27}$$

where $L_M$ is the maximum distance between the electron and the source. In this case Eq. (3) can be regarded as a good approximation, and the main transfer of the energy from the electron to the source is expected to be made through the electric field associated with the change in the four potential of the moving electron. Let us subdivide the stationary current of the source into a number of loops $C_\lambda$ each of which has a very small cross section.[23),1)] Then, the work dW done by the induced voltage at the loop $C_\lambda$ during the time dt is

$$\begin{aligned} dW_\lambda &= \oint_{C_\lambda} \mathbf{e}(\mathbf{r}_\lambda) \cdot I_\lambda d\mathbf{l}_\lambda \delta t = dt \cdot \oint_{C_\lambda} -\frac{1}{c}\frac{\partial \mathbf{a}(\mathbf{r}_\lambda)}{\partial t} \cdot I_\lambda d\mathbf{l}_\lambda \\ &= dt \cdot \frac{d}{dt}\left[\frac{e\mathbf{v}}{4\pi c^2} \cdot \oint_{C_\lambda} \frac{I_\lambda d\mathbf{l}_\lambda}{|\mathbf{r} - \mathbf{r}_\lambda|}\right] = dt \cdot \frac{d}{dt}\left[\frac{e\mathbf{v}}{c} \cdot \mathbf{A}_\lambda(\mathbf{r})\right] \end{aligned} \tag{28}$$

Here $\mathbf{e}(\mathbf{r}_\lambda)$ is the electric field induced by the change of the vector potential $\mathbf{a}(\mathbf{r})$. Then the total work given to the source, dW, is

$$dW = \sum_\lambda dW_\lambda = dt \cdot \frac{d}{dt}\left[\frac{e\mathbf{v}}{c} \cdot \mathbf{A}(\mathbf{r})\right] \tag{29}$$

Here $\mathbf{A}(\mathbf{r})$ is the vector potential of the source at the location of the electron. Namely

$$d\left(-\frac{e}{c}\mathbf{v} \cdot \mathbf{A} + W\right) = 0 \tag{30}$$

In this way, we have two constants of the motion, i.e., the Hamiltonian of the electron

$$\mathcal{H} = mc^2\sqrt{1 + \frac{(\mathbf{p} + \frac{e}{c}\mathbf{A})^2}{m^2c^2}} - e\varphi \tag{31}$$

and the total energy of the system as represented by Eq. (19) or (7).

Now let us analyze the second case in which

$$\Delta L < L_M \tag{32}$$

Let us assume that an electron in a cyclotron motion collides with a neutral molecule. Let us assume further a typical elastic case in which the electron and the molecule have reversed their momenta and there is no change in the energy of the molecule. Then the electromagnetic signal containing the information that the electron has reversed its momentum is confined in a spherical shell with thickness $\Delta L$ which propagates outwards with the speed of light from the colliding place. Shortly after the collision, this travelling information arrives at the coil and will react to the current of the coil, introducing a change either in the current or in the stored energy of the source of the current. We can solve this problem completely rigorously.

It is to be noted that the direction of the propagation of the spherical shell and the direction of the energy flow are not necessarily identical[24] and we know that the amount of energy which would be radiated into the infinite space is quite small[11]. In this example, the associated magnetic energy change of the system for the imaginary stationary state is $2e\mathbf{v}_0 \cdot \mathbf{A}/c$, which can be either positive or negative and could be as high as several tens of electron volts. At the time of collision, however, this change had to be compensated by the associated electromagnetic fields state arround the electron and this information with a negative or positive energy has to propagate with the speed of light over the entire system to realize a new stationary state. The conservation of total energy in the form of Eq. (19) can be realized only after these electromagnetic information has covered the entire system.

## § 5. Correct and Incorrect Understandings of the Magnetic Energy and An Essential Advantage of the Lagrangian Formalism over the Hamiltonian Formalism for a Macroscopically Inhomogeneous System

From Eq. (7), the total Lagrangian, momenta, and energy of a system in a quasi-stationary state can be defined as[20]

$$L = -\sum_i m_i c^2 + \sum_i \frac{m_i}{2} v_i^2 + \sum_i \frac{m_i v_i^4}{8c^2} - \sum_{i>j} \frac{q_i q_j}{4\pi r_{ij}}$$

$$+ \sum_{i>j} \frac{q_i q_j (\mathbf{v}_i \cdot \mathbf{v}_j)}{4\pi r_{ij} c^2} - \sum_{i>j} \frac{q_i q_j \left[(\mathbf{v}_i \cdot \mathbf{v}_j) + (\mathbf{v}_i \hat{\mathbf{r}}_{ij})(\mathbf{v}_j \cdot \hat{\mathbf{r}}_{ji})\right]}{8\pi r_{ij} c^2} \tag{33}$$

$$p_i = \frac{\partial L}{\partial v_i} = m_i v_i + \frac{m_i v_i^2}{2c^2} v_i + \sum_{j \neq i} \frac{q_i q_j v_j}{4\pi r_{ij} c^2}$$

$$- \sum_{j \neq i} \frac{q_i q_j \left[v_j + \hat{r}_{ij} (v_j \cdot \hat{r}_{ji})\right]}{8\pi r_{ij} c^2} \tag{34}$$

$$\frac{dp_i}{dt} = \frac{\partial L}{\partial r_i} \tag{35}$$

$$U = \sum_i m_i c^2 + \sum_i \frac{m_i}{2} v_i^2 + \sum_i \frac{3 m_i v_i^4}{8c^2} + \sum_{i > j} \frac{q_i q_j}{4\pi r_{ij}}$$

$$+ \sum_{i > j} \frac{q_i q_j (v_i \cdot v_j)}{4\pi r_{ij} c^2} - \sum_{i > j} \frac{q_i q_j \left[(v_i \cdot v_j) + (v_i \cdot \hat{r}_{ij})(v_j \cdot \hat{r}_{ji})\right]}{8\pi r_{ij} c^2} \tag{36}$$

Here, i's designate many electrons and nuclei. We shall call Eq. (36) as the Lagrangian energy. As has been mentioned in Eq. (21), the term

$$\sum_{j \neq i} \frac{q_i q_j v_j}{4\pi r_{ij} c^2} = \frac{q_i}{c} A_i(r_i) \tag{37}$$

of Eq. (34) cannot be small in a usual situation. The order estimation will be

$$m_i v_i : \frac{q_i}{c} A_i(r_i) = v_i : \frac{e}{mc} \frac{H \cdot r}{2} = v_i : 5 \times 10^{6 \sim 7} \text{ m/sec} \tag{38}$$

at the surface of a specimen with a radius of 0.5 cm and with the magnetic field of 100 ~ 1000 Oe. Here we can assume $v_i \lesssim 3 \times 10^{3 \sim 5}$ m/sec, and, as has ben mentioned, for a quasi-stationary current at a long distance

$$\sum_{j \neq i} \frac{q_j \left[v_j + \hat{r}_{ij}(v_j \cdot \hat{r}_{ji})\right]}{8\pi r_{ij} c^2} = 0 \tag{39}$$

can be assumed. Then we can adopt a perturbation theory to Eq. (34) to get $v_i$. The result is

$$v_i = D_i [p_\xi] + D_i \Big[ - \frac{m_\eta \{D_\eta [p_\xi]\}^2}{2c^2} D_\eta [p_\xi]$$

$$+ \sum_{j \neq \eta} \frac{q_\eta q_j (D_j [p_\xi] + \hat{r}_{\eta j} \{D_j [p_\xi] \cdot \hat{r}_{j\eta}\})}{8\pi r_{\eta j} c^2} \Big] \tag{40}$$

where

$$
D_i\,[x_\xi] = \frac{\begin{array}{cccccc} 1 & 2 & \cdot\cdot\, i & \cdots\cdots & & N \\ \end{array}\\ \begin{vmatrix} m_1 & \dfrac{q_1 q_2}{4\pi r_{12} c^2} & \cdot\cdot\, x_1 \cdots\cdots & \dfrac{q_1 q_N}{4\pi r_{1N} c^2} \\ \dfrac{q_2 q_1}{4\pi r_{21} c^2} & m_2 & \cdot\cdot\, x_2 \cdots\cdots & \dfrac{q_2 q_N}{4\pi r_{2N} c^2} \\ \vdots & \vdots & \vdots & \vdots \\ \dfrac{q_N q_1}{4\pi r_{N1} c^2} & \dfrac{q_N q_2}{4\pi r_{N2} c^2} & \cdot\cdot\, x_N \cdots\cdots & m_N \end{vmatrix}}{\begin{vmatrix} m_1 & \dfrac{q_1 q_2}{4\pi r_{12} c^2} & \cdots\cdots & \dfrac{q_1 q_N}{4\pi r_{1N} c^2} \\ \dfrac{q_2 q_1}{4\pi r_{21} c^2} & m_2 & \cdots\cdots & \cdot \\ \vdots & \vdots & & \vdots \\ \dfrac{q_N q_1}{4\pi r_{N1} c^2} & \cdots\cdots & \cdots\cdots & m_N \end{vmatrix}} \tag{41}
$$

The true Hamiltonian of the system will be obtained by inserting Eq. (41) into Eq. (36). When we take only the principal terms, we get

$$
\begin{aligned}
\mathcal{H}^{(0)} &= \sum_i m_i c^2 + \sum_i \frac{m_i}{2} \{D_i\,[p_\xi]\}^2 + \sum_{i>j} \frac{q_i q_i}{4\pi r_{ij}} \\
&\quad + \sum_{i>j} \frac{q_i q_j \{D_i\,[p_\xi] \cdot D_j\,[p_\xi]\}}{4\pi r_{ij} c^2} \\
&= \sum_i m_i c^2 + \sum_i \frac{p_i^2}{2m_i} + \sum_{i>j} \frac{q_i q_j}{4\pi r_{ij}} - \sum_{i>j} \frac{q_i q_j (D_i \cdot D_j)}{4\pi r_{ij} c^2} \\
&\quad - \sum_i \frac{1}{2m_i} \Big( \frac{q_i}{c} \sum_{j \neq i} \frac{D_j}{4\pi r_{ij} c} \Big)^2
\end{aligned} \tag{42}
$$

which is quite different from the conventional Hamiltonian.[25)]

As has been shown partly in § 4, we can verify easily that the conventional Hamiltonian is an effective Hamiltonian which is valid only for the kinematical description of the electron. Even when the radiation terms are included,[24),25)] it cannot include the magnetic interaction energy between the source of the quasi-static magnetic field and the electrons, so that it is inadequate for the treatment of statistical thermodynamics of the system.[26),27)]

For our problem, therefore we must use Eq. (42) as the basic Hamiltonian. Now, besides the expected mathematical difficulty, we shall point out another essential difficulty for the Hamiltonian of a macroscopically inhomogeneous system. We know from the argument of § 4, that the Lagrangian treatment is only approximately correct because of the presence of finite velocity for the propagation of electromagnetic informations. In the case of Lagrangian treatment, however, the differentiation in the treatment should be made at constant $\mathbf{v}_j$, which corresponds to fix the vector potential arround $\mathbf{r}_i$, and well acceptable physically. In the case of Hamiltonian treatment, however, the differentiation should be made at constant $\mathbf{p}_\xi$, which has no physical correspondence, and the effect of all the errors due to the finite velocity of the information transfer may accumulate in a worse way. In our macroscopically inhomogeneous system, the effect of the presence of surface boundary is crucial and we don't know how this effect will appear in Hamiltonian treatment. Therefore, from now on we shall place the primary importance to the Lagrangian treatment.[28)]

## § 6. Preliminary Consideration on the Thermodynamical Treatment of a Perfect Conductor

What we hope to verify is the fact that when a material can sustain a persistent current and can be called as a perfect conducter, then the material should show the Meissner effect automatically. Let us postulate a classical electron gas in a rigid ion core lattice. The effect of the rigid lattice will be represented by a rigid positive ion core potential $\varphi^{+}(\mathbf{r})$, which, after combining the effect of the majority of the electrons, will eventually be assumed to be uniform.

The subject of our study is a simply connected classical electron gas $C_2$ in a magnetic field $\mathbf{H}_1$. In order to define the total system accurately, another doubly connected classical electron gas $C_1$, which keeps a persistent current, will be assumed as the source of the magnetic field.

Now, we shall pick up four important characters of the system. They are as follows.
1). We assume that any electric current distribution $\mathbf{j}_2(\mathbf{r})$ in $C_2$, which satisfies

$$\nabla \cdot \mathbf{j}_2(\mathbf{r}) = 0 \tag{43}$$

can be metastable, on the basis of the Maxwell equations. When $\mathbf{j}_2(\mathbf{r})$ and $\mathbf{H}(\mathbf{r})$ coexist, we have to expect the presence of a magnetic Lorentz force, but this force is balanced by an electric Lorentz force, which is introduced by an induced extremely small displacement of electrons (such as $10^{-10}$ Å !). On the other hand the time constant for the electrons to arrive at a local equilibrium state in a local small volume dV, will be very short such as

$$\tau = \frac{2\text{Å}}{v} = \frac{2 \times 10^{-10}\,\text{m}}{c \times 10^{-(3\sim 5)}} = 7 \times 10^{-(14\sim 16)}\ \text{sec.} \tag{44}$$

The change of $\mathbf{j}_2(\mathbf{r})$ will be made through thermal fluctuations in terms of the Maxwell-Lorentz equations and will need a relaxation time which is longer than Eq. (44) probably by a factor of at least more than one order of magnitude..

2). From Eq. (36), the effectively changeable part of the total energy of the system, which is denoted by U, is

$$U = \sum_i \frac{m}{2} v_i^2 + \frac{1}{2}\sum_i \{-e\,[\sum_j \frac{-e}{4\pi r_{ij}} + \varphi^+(\mathbf{r}_i)]\} + \frac{1}{2}\sum_i \{-\frac{e\mathbf{v}_i}{c}\cdot[\sum_j \frac{-e\mathbf{v}_j}{4\pi r_{ij} c}]\}$$

$$= \sum_i \frac{m}{2}[\mathbf{v}_i - \mathbf{v}_D(\mathbf{r}_i)]^2 + \sum_i \frac{m}{2}[\mathbf{v}_D(\mathbf{r}_i)]^2 + \frac{1}{2}\sum_i \{-\frac{e\mathbf{v}_i}{c}\cdot \mathbf{A}(\mathbf{r}_i)\} \tag{45}$$

Here

$$\mathbf{v}_D(\mathbf{r}) = (\sum_{i\ \text{in}\ dV} \mathbf{v}_i) / \sum_{i\ \text{in}\ dV} 1 \tag{46}$$

is a drift velocity at $\mathbf{r}$, and, assuming a uniform distribution of electrons in $C_1$ and $C_2$, we put the average electric potential as zero. We have used the macroscopic vector potential $\mathbf{A}(\mathbf{r})$ in stead of the individual sum. Because when a usual macroscopic vector potential is considered, the ratio of this term to the microscopic vector potential of a single electron at 2 Å apart is quite small, e.g.,

$$\frac{e \times c \times 10^{-(2\sim 4)}}{4\pi\,(2 \times 10^{-10})\,c} : \frac{1}{2} H \times R = 2 \times 10^{-(7\sim 9)} : 2 \times 10^{-2} \tag{47}$$

at the location 5 mm from the axis of symmetry of a magnetic field of 100 Oe with cylindrical symmetry. Introducing the average electron density $n(\mathbf{r})$, we obtain from Eq. (45)

$$U = \sum_i \frac{m}{2}[\mathbf{v}_i - \mathbf{v}_D(\mathbf{r}_i)]^2 + \iiint_{V_1+V_2} \frac{n(\mathbf{r})\,m}{2}[\mathbf{v}_D(\mathbf{r})]^2\, dV + \iiint_{V_1+V_2} \frac{\mathbf{j}(\mathbf{r})}{2c}\cdot \mathbf{A}(\mathbf{r})\, dV$$

$$= \iiint_{V_1 + V_2} \frac{n\,m}{2} \overline{\overline{(v_i - v_D)^2}}\, dV + \iiint_{V_1 + V_2} [\frac{m}{2n\,e^2} j^2 + \frac{1}{2c} \mathbf{j} \cdot \mathbf{A}\,]\, dV$$

$$= U_{kT} + U_{kD} + U_m \ . \qquad (48)$$

Here $V_1$ and $V_2$ are the volumes of $C_1$ and $C_2$, respectively and the current density is

$$\mathbf{j}(\mathbf{r}) = -\,n(\mathbf{r})\,e\ \mathbf{v}_D\,(\mathbf{r}) \qquad (49)$$

The first, second, and third terms of Eq. (45) can be regarded as the thermal and drift parts of the kinetic energy and the magnetic energy of the electrons, respectively.

We conclude further that $n(\mathbf{r}) = n_2$ is constant very accurately over the entire volume. This can be verified quantitatively in the following way. Suppose $C_2$ has been electrically charged up with additional electrons. Then our statistical thermodynamical calculation can show that the additional electrons must be on the surface of $C_2$, and the distribution of this charge at the surface can be represented as,

$$\rho\,(\zeta) = \rho_0 \exp(-\frac{\zeta}{\Lambda'}) \qquad (50)$$

in which $\zeta$ is a cartesian coordinate, whose axis is normal to the surface. The penetration depth

$$\Lambda' = \sqrt{\frac{kT}{\bar{\bar{n}}_2 e^2}} \qquad (51)$$

in which $\bar{\bar{n}}_2$ is the average density of the electrons. At $T = 10^{(-1 \sim +3)}{}^\circ K$

$$\Lambda' = 10^{-(13 \sim 11)} m = 10^{-(3 \sim 1)}\ \text{Å} \qquad (52)$$

which is an awfully small distance. When $C_2$ is a sphere of the radius 1 cm which is charged up to $10^2$ Volt, the mean distance of the additional electrons at the surface is about $1\ \mu =$ 10,000 Å[7]) which is very large. These situations conclude that when the dominant interaction is magnetic and is very weak, the electrostatic interactions are so strong as to keep $n_2$ rigorously constant over the entire volume.

3). We assume that, when our system $C_1 + C_2$ is close to its equilibrium, the variation of the entropy of the system depends only on the structure of the integrand of the first term of Eq. (45). and is almost independent of $\mathbf{j}(\mathbf{r})$.

4). When we consider a local small volume V in $C_2$, we can classify important energy transfer processes in the following way.

A Transfer of thermal kinetic energy at the surface.

A–I, through collisions and mass actions of electrons at the surface. The role of the drift velocity $v_D$ will be small, because the ratio will be

$v_i : v_D = 3 \times 10^{5\sim3} : 3 \times 10^{(1\sim-1)}\ (\mathrm{m\,s^{-1}})$

A–II, emission and absorption of Maxwell–Lorentz electromagnetic thermal radiation through the surface.

B Macroscopic electromagnetic energy transfer through the Maxwell equation. It is expressed by

$$-\iint_S c\mathbf{E} \times \mathbf{H} \cdot d\mathbf{S} = \iiint_V \left[\frac{\partial}{\partial t}\left(\frac{\mathbf{E}^2 + \mathbf{H}^2}{2}\right) + \mathbf{E} \cdot \mathbf{j}\right] dV \ . \tag{53}$$

Here S is the surface of V. Component processes are:

B–I. Going out or coming in of the macroscopic electromagnetic energy.

B–II. Change of the macroscopic electric and magnetic energies in V.

B–III. Transformation of macroscopic electromagnetic energy into non-electromagnetic energy $U_{kT}$ or $U_{kD}$ of Eq. (48).

B–IV. Transformation of non-electromagnetic energy $U_{kT}$ or $U_{kD}$ into the macroscopic electromagnetic energy.

Processes B-III and IV are represented by the term of $\mathbf{E} \cdot \mathbf{j}$, which is most important and most complicated.

Let us analyze Eq. (53) with an emphasis on $\mathbf{E} \cdot \mathbf{j}$ term. We assume that

$$\left.\begin{array}{ll} t \leq t_0 & \mathbf{j}_2(\mathbf{r}, t) = \mathbf{j}_2(\mathbf{r}) \\ t_0 \leq t \leq t_0 + \delta t_0 & \mathbf{j}_2(\mathbf{r}, t) = \mathbf{j}_2(\mathbf{r}) + \delta \mathbf{j}_2(\mathbf{r}, t) \\ t \geq t_0 + \delta t_0 & \mathbf{j}_2(\mathbf{r}, t) = \mathbf{j}_2(\mathbf{r}) + \delta \mathbf{j}_2(\mathbf{r}) \end{array}\right\} \tag{54}$$

This means that a metastable current distribution $\mathbf{j}_2(\mathbf{r})$ is transforming towards another metastable current distribution $\mathbf{j}_2(\mathbf{r}) + \delta\mathbf{j}_2(\mathbf{r})$ through a physical action, which could be dynamical or thermal and will be described by the Maxwell–Lorentz electromagnetism. Then we get from

Eq. (52)

$$-\int_{t_0}^{t_0+\delta t}\iint_S c\mathbf{E}\times\mathbf{H}\cdot d\mathbf{S}\,dt = \iiint_V [\,\delta\left(\frac{\mathbf{H}^2}{2}\right)+\int_{t_0}^{t_0+\delta t}\mathbf{E}(\mathbf{r},t)\cdot\mathbf{j}_2(\mathbf{r},t)\,dt]\,dV \quad (55)$$

Since $\Delta\cdot\delta\mathbf{j}_2(\mathbf{r}) \neq 0$, $\delta\mathbf{j}_2(\mathbf{r})$ should construct a small current loop. However, when $\mathbf{j}_2(\mathbf{r}) = 0$, the introduction of $\delta\mathbf{j}_2(\mathbf{r})$ is a slight shift of $\mathbf{j}_2(\mathbf{r})$. Now, when we assume that dV indicates the smallest volume that is still large microscopically,

$$\mathbf{j}_2(\mathbf{r},t) = -e\,[\sum_{i\text{ in }dV}\mathbf{v}_i(t)\,]/dV = -en_2\mathbf{v}_D(\mathbf{r},t) \quad (56)$$

and the momentum in dV is

$$\mathbf{p}_{dV} = m\sum_{i\text{ in }dV}\mathbf{v}_i = m\,[\,\mathbf{v}_D(\mathbf{r})+\delta\mathbf{v}_D(\mathbf{r},t)\,]\,n_2\,dV\ . \quad (57)$$

From electrodynamics, we must expect the presence of a Lorentz force $\mathbf{f}_2(\mathbf{r},t)$

$$\mathbf{f}_2(\mathbf{r},t) = -ne\,\mathbf{E}(\mathbf{r},t)+\frac{\mathbf{j}_2(\mathbf{r},t)}{c}\times\mathbf{H}(\mathbf{r},t) \quad (58)$$

As we have mentioned already, since we have assumed that $\mathbf{j}_2(\mathbf{r})$ and $\mathbf{j}_2(\mathbf{r})+\delta\mathbf{j}_2(\mathbf{r})$ are stable, we have to assume at

$$t\leq t_0 \qquad \mathbf{E}(\mathbf{r}) = -\frac{\mathbf{v}_D(\mathbf{r})}{c}\times\mathbf{H}(\mathbf{r})$$

$$t\geq t_0+\delta t_0 \qquad \mathbf{E}(\mathbf{r})+\delta\mathbf{E}(\mathbf{r}) = -\frac{[\mathbf{v}_D(\mathbf{r})+\delta\mathbf{v}_D(\mathbf{r})\,]}{c}\times[\,\mathbf{H}(\mathbf{r})+\delta\mathbf{H}(\mathbf{r})\,] \quad (59)$$

$$\delta\mathbf{E}(\mathbf{r}) = -\frac{\mathbf{v}_D(\mathbf{r})}{c}\times\delta\mathbf{H}(\mathbf{r})-\frac{\delta\mathbf{v}_D(\mathbf{r})}{c}\times\mathbf{H}(\mathbf{r})-\frac{\delta\mathbf{v}_D(\mathbf{r})}{c}\times\delta\mathbf{H}(\mathbf{r})\ . \quad (60)$$

$\mathbf{E}(\mathbf{r})$ and $\delta\mathbf{E}(\mathbf{r})$ are extremely small electric fields. Since these electric fields have no work because $\mathbf{E}\cdot\mathbf{j} = 0$, we can neglect the action of these fields. Then

$$\frac{d}{dt}m\sum_{i\text{ in }dV}\delta\mathbf{v}_i(\mathbf{r}_i,t) = -\sum_{i\text{ in }dV}e[\,\mathbf{E}(\mathbf{r}_i)+\delta\mathbf{E}(\mathbf{r}_i,t)]$$

$$-\sum_{i\text{ in }dV}e\,\frac{[\mathbf{v}_i(\mathbf{r}_i)+\delta\mathbf{v}_i(\mathbf{r}_i,t)]}{c}\times[\mathbf{H}(\mathbf{r}_i)+\delta\mathbf{H}(\mathbf{r}_i,t)]+\delta\mathbf{p}_T(\mathbf{r},t) \quad (61)$$

$$\frac{d}{dt} m\, n_2\, \delta \mathbf{v}_D (\mathbf{r}, t) = - e n_2 \; [\mathbf{E}(\mathbf{r}) + \delta \mathbf{E}(\mathbf{r}, t)]$$

$$- e n_2 \frac{[\mathbf{v}_D (\mathbf{r}) + \delta \mathbf{v}_D (\mathbf{r}, t)]}{c} \times [\mathbf{H}(\mathbf{r}) + \delta \mathbf{H} (\mathbf{r}, t)] + \delta \mathbf{P}_T(\mathbf{r}, t) \tag{62}$$

$\delta \mathbf{P}_T(\mathbf{r}, t) = \delta \mathbf{p}_T(\mathbf{r}, t)/dV$ indicates the action of or to the random thermal motion of the electrons. Multiplying $[\mathbf{v}_D (\mathbf{r}) + \delta \mathbf{v}_D (\mathbf{r}, t)]$, we get

$$m n_2 \cdot \frac{d}{dt} \frac{\left\{ \mathbf{v}_D(\mathbf{r}, t) \right\}^2}{2} = -e n_2 \; [\mathbf{E}(\mathbf{r}) + \delta \mathbf{E} (\mathbf{r}, t)] \cdot \mathbf{v}_D (\mathbf{r}, t) + \delta \mathbf{P}_T (\mathbf{r}, t) \cdot \mathbf{v}_D (\mathbf{r}, t)$$

$$= \mathbf{j}_2 (\mathbf{r}, t) \cdot \mathbf{E} (\mathbf{r}, t) + \mathbf{v}_D (\mathbf{r}, t) \cdot \delta \mathbf{P}_T (\mathbf{r}, t) \; . \tag{63}$$

We regard Eq. (63) as the most fundamental equation of the problem. Owing to the very rapid stochastic motions in a magnetic field, a group of electrons can have a possibility to create $\delta \mathbf{j}(\mathbf{r}, t)$ or $\delta \mathbf{v}_D(\mathbf{r}, t)$ spontaneously with a consumption of their thermal kinetic energies, provided that the intial state is not in equilibrium. Then from Eq. (55), we get at $t = t_0 + \delta t$

$$-\int_{t_0}^{t_0 + \delta t} \iint_S c\mathbf{E} \times \mathbf{H} \cdot d\mathbf{S}\, dt = \iiint_V [\, \delta (\frac{\mathbf{H}^2}{2}) + \frac{m}{n_2 e^2} \delta (\frac{j_2^2}{2}) \,] \; dV$$

$$- \iiint_V \int_{t_0}^{t_0 + \delta t} \mathbf{v}_D \cdot \delta \mathbf{P}_T dt\, dV \; . \tag{64}$$

Therefore, the electromagnetic energy transfered from (or into) the volume V, $\delta U_{trans}$, is

$$\delta U_{trans} = - \iiint_V \; [\mathbf{H} \cdot \delta \mathbf{H} + \frac{m}{ne^2} \mathbf{j}_2 \cdot \delta \mathbf{j}_2 + \frac{(\delta \mathbf{H})^2}{2} + \frac{m}{n_2 e^2} \frac{(\delta \mathbf{j}_2)^2}{2}] \; dV - \iiint_V \delta u_{kT} \; dV \; . \tag{65}$$

In which $\delta u_{kT}$ indicates the unit volume change of the local kinematical thermal energy of the electrons. By assuming that $\delta \mathbf{j}_2$ and $\delta t$ are small, we have neglected the presence of process A in Eq. (65).

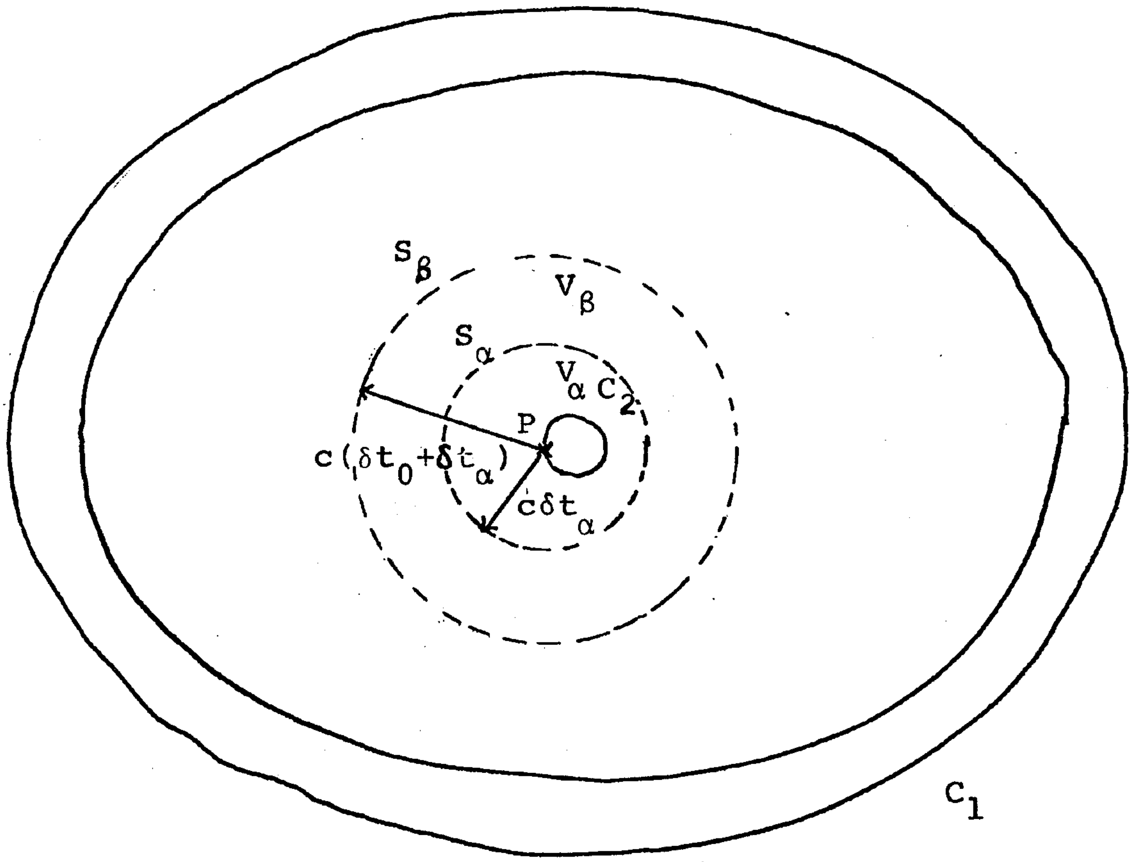
Let us extend our analysis further with assuming that $C_1$ is very large and located at a very distant place. As is shown in Fig. 1, let us assume that $\delta \mathbf{j}_2$ $(\mathbf{r}, t)$ starts at $t = t_0$ at the point P in $C_2$ and it ends in $C_2$ completely at $t = t_0 + \delta t_0$. We take a surface $S_\alpha$ at $t = t_0 + \delta t_0 + \delta t_\alpha$, which is located in an empty space with a radius $c\delta t_\alpha$ from P. As shown in Fig. 1 another cocentered sphere $S_\beta$ can be taken at $t = t_0 + \delta t_0 + \delta t_\alpha$, for which the radius is $c(\delta t_0 + \delta t_\alpha)$. $S_\beta$ indicates the wave front of the considered change of $\delta \mathbf{j}_2$ and we can conclude that all the transient electromagnetic informations are located in a space bounded by $S_\beta$ and $S_\alpha$, i. e., $V_\beta - V_\alpha$. Here $V_\beta$ and $V_\alpha$ are the volumes inside of $S_\beta$ and $S_\alpha$, respectively, and we can assume that there is no transient electromagnetic fields inside of $S_\alpha$ or in $V_\alpha$. Then from Eqs. (53), (64) and (65), we get rigorously an electromagnetic energy relaton at $S_\alpha$ and in $V_\alpha$ as

$$\begin{aligned}\delta U^\alpha_{\text{trans}} &= \int_{t_0}^{t_0 + \delta t_0 + \delta t_\alpha} \iint_{S_\alpha} c\mathbf{E} \times \mathbf{H} \cdot d\mathbf{S}\ dt \\ &= - \iint_{S_\alpha} \mathbf{A}^0 \times \delta \mathbf{H}^\alpha_2 \cdot d\mathbf{S} - \iiint_{V_2} \left(\mathbf{A}^0 + \frac{mc}{n_2 e^2}\, \mathbf{j}^0_2\right) \cdot \frac{\delta \mathbf{j}^\alpha_2}{c}\, dV \\ &\quad - \iiint_{V_\alpha} \left[\frac{(\delta \mathbf{H}^\alpha_2)^2}{2} + \frac{m}{n_2 e^2} \frac{(\delta \mathbf{j}^\alpha_2)^2}{2}\right] dV - \iiint_{V_2} \delta u^\alpha_{KT}\, dV \end{aligned} \tag{66}$$

Here, $\mathbf{H}^0$, $\mathbf{A}^0$, and $\mathbf{j}^0_2$ indicate the values of the functions at $t \leq t_0$ and $\delta \mathbf{H}^\alpha_2$ and $\delta \mathbf{j}^\alpha_2$ indicates the values at $t = t_0 + \delta t_0 + \delta t_\alpha$. Care should be taken that, although $\delta \mathbf{H}^\alpha_2$ will decay as $1/r^3$ in $V_\alpha$, the first surface integral term of the last equation of Eq. (66) can not be put as zero, because not $\mathbf{A}^0$ but $\nabla \times \mathbf{A}^0 = \mathbf{H}^0$ has a significance in this equation.

When we see the same phenomena from the outside space of $V_\alpha$, we get

$$\begin{aligned}\delta U^\alpha_{\text{trans}} &= \iiint_{\infty - V_\alpha} \left[\delta \left(\frac{\mathbf{H}^2}{2}\right) + \frac{(\delta \mathbf{E})^2}{2}\right] dV \\ &= - \iint_{S_\alpha} \mathbf{A}^0 \times \delta \mathbf{H}^\alpha_2 \cdot d\mathbf{S} + \iiint_{V_\beta - V_\alpha} \mathbf{A}^0 \cdot \frac{1}{c} \frac{\partial \delta \mathbf{E}^\alpha}{\partial t}\, dV\end{aligned}$$




at $t = t_0 + \delta t_0 + \delta t_\alpha$

Fig. 1 A simplest illustration of a transient travelling electromagnetic fields.

$$+\iiint_{V_\beta - V_\alpha} [\frac{(\delta \mathbf{H}^\alpha)^2}{2} + \frac{(\delta \mathbf{E}^\alpha)^2}{2}] \ dV \qquad (67)$$

On the other hand, if we observe $\delta U_{trans}$ at the surface infinitely remote and in the time interval $[t_0, t_0 + \infty]$,

$$\delta U^{\infty}_{trans} = \int_{t_0}^{t_0+\infty} \iint_{S_\infty} c\mathbf{E} \times \mathbf{H} \cdot d\mathbf{S} \ dt$$

$$= - \iiint_{V_\infty} [\delta (\frac{\mathbf{H}^2 + \mathbf{E}^2}{2}) + \int_{t_0}^{t_0+\infty} \mathbf{E}\cdot\mathbf{j} \ dt] \ dV \qquad (68)$$

When we assume that $\mathbf{j}_1^0$ ($\mathbf{r}$) is perfectly rigid, we have

$$\delta U^r_R = \delta U^{\infty, r}_{trans} = - \iiint_{V_\infty} [\mathbf{H}^0 \cdot \delta \mathbf{H} + \frac{(\delta \mathbf{H})^2}{2}] \ dV - \int_{t_0}^{\infty} \iiint_{V_1} \delta \mathbf{E} \cdot \mathbf{j}_1^0 \ dV$$

$$- \int_{t_0}^{t_0+\delta t_0} \iiint_{V_2} \delta \mathbf{E} \cdot \mathbf{j}_2 \ dV \ dt$$

$$= - \delta W^r_1 - \iiint_{V_2} \mathbf{A}^0 \cdot \frac{\delta \mathbf{j}_2^\alpha}{c} dV - \iiint_\infty \frac{(\delta \mathbf{H})^2}{2} dV - \int_{t_0}^{t_0+\delta t_0} \iiint_{V_2} \delta \mathbf{E}\cdot\mathbf{j}_2 \ dVdt$$

$$(69)$$

Here, $\delta W^r_1$ is the work done to $C_1$ by the travelling electric field $\delta \mathbf{E}$ and $\delta U^r_R$ is the radiation energy which has escaped to infinity.

Then, the total energy $\delta U$ which is transfered from $C_2$ to the other systems is

$$\delta U = \delta W^r_1 + \delta U^r_R$$

$$= - \iiint_{V_2} \mathbf{A}^0 \cdot \frac{\delta \mathbf{j}_2^\alpha}{c} \ dV - \iint_\infty \frac{(\delta \mathbf{H})^2}{2} \ dV - \int_{t_0}^{t_0+\delta t_0} \iiint_{V_2} \delta \mathbf{E} \cdot \mathbf{j}_2 \ dV \ dt \ .$$

$$(70)$$

From Eq. (66), we see that the surface integral

$$\iint_{S_\alpha} \mathbf{A}^0 \times \delta \mathbf{H}_2^\alpha \cdot d\mathbf{S} = \iiint_{V_\alpha} \nabla \cdot (\mathbf{A}^0 \times \delta \mathbf{H}_2^\alpha) \ dV \qquad (71)$$

has no action to $\delta U$.

It is not difficult to obtain

$$\iiint_{V_\beta - V_\alpha} \mathbf{A}^0 \cdot \frac{1}{c}\frac{\partial \delta \mathbf{E}^\alpha}{\partial t}\, dV = -\iiint_{V_2} \frac{\mathbf{A}_1^0 \cdot \delta \mathbf{j}_2^\alpha}{c}\, dV \tag{72}$$

by a vector analysis. Then from Eqs. (66), (67) and (70), we get

$$\delta U = -\iiint_{V_2} \frac{\mathbf{A}_1^0 \cdot \delta \mathbf{j}_2^\alpha}{c}\, dV + \iiint_{V_\beta - V_\alpha} \left[\frac{(\delta \mathbf{E}^\alpha)^2}{2} + \frac{(\delta \mathbf{H}^\alpha)^2}{2}\right] dV$$

$$-\iiint_{\infty - V_\alpha} \frac{(\delta \mathbf{H})^2}{2}\, dV\ . \tag{73}$$

On the other hand $\delta W_1^r$ can be calculated directly as

$$\delta W_1^r = \int_{t_0}^{\infty} \iiint_{V_1} \delta \mathbf{E} \cdot \mathbf{j}_1^0\, dV\, dt = \int_{t_0}^{\infty} \iint_{V_1} \left(-\frac{1}{c}\frac{\partial \delta \mathbf{A}}{\partial t} - \nabla \delta\varphi\right) \cdot \mathbf{j}_1^0\, dV\, dt$$

$$= -\iiint_{V_2} \frac{\mathbf{A}_1^0 \cdot \delta \mathbf{j}_2^\alpha}{c}\, dV = \iiint_{V_\beta - V_\alpha} \mathbf{A}^0 \cdot \frac{1}{c}\frac{\partial \delta \mathbf{E}^\alpha}{\partial t}\, dV \tag{74}$$

Therefore from Eq. (73)

$$\delta U_R^r = \iiint_{V_\beta - V_\alpha} \left[\frac{(\delta \mathbf{E}^\alpha)^2}{2} + \frac{(\delta \mathbf{H}^\alpha)^2}{2}\right] dV - \iiint_{\infty - V_\alpha} \frac{(\delta \mathbf{H})^2}{2}\, dV \tag{75}$$

which is interpreted as a thermal radiation. From Eqs. (74), (72) and (66).

$$\delta W_1^r = -\iiint_{V_2} \frac{\mathbf{A}_1^0 \cdot \delta \mathbf{j}_2^\alpha}{c}\, dV = \iiint_{V_\beta - V_\alpha} \mathbf{A}^0 \cdot \frac{1}{c}\frac{\partial \delta \mathbf{E}^\alpha}{\partial t}\, dV \tag{76}$$

$$= -\iiint_{V_2} \left(\mathbf{A}^0 + \frac{mc}{n_2 e^2}\, \mathbf{j}_2^0\right) \cdot \frac{\delta \mathbf{j}_2^\alpha}{c}\, dV - \iiint_{V_\alpha} \left[\frac{(\delta \mathbf{H}^\alpha)^2}{2} + \frac{m(\delta \mathbf{j}_2^\alpha)^2}{2 n_2 e^2}\right] dV$$

$$- \iiint_{V_\beta - V_\alpha} \left[\frac{(\delta \mathbf{E}^\alpha)^2}{2} + \frac{(\delta \mathbf{H}^\alpha)^2}{2}\right] dV - \iiint_{V_2} \delta u_{kT}^\alpha\, dV\ . \tag{77}$$

One of the essential character of the present problem is that $C_1$ can see $C_2$ only as a macroscopic electromagnetic subject, so that only $\delta j_2^{\alpha}$ can have an action to $C_1$ as shown in Eq. (76), whereas there are complicated interactions between $U_{kT}$, $U_{kD}$ and $U_{e.m.}$ in $C_2$, which are represented in Eq. (77). It is obvious from Eq. (76), that, in order to get positive $\delta W_1^r$, $\delta j_2^{\alpha}$ must be diamagnetic, since

$$-\iiint_{V_2} \frac{A_0^1 \cdot \delta j_2^{\alpha}}{c}\, dV = -\sum_i \frac{\Delta I_i}{c} \oint_{C_i} A_1^0 \cdot d\ell_i = -\sum_i \frac{\Delta I_i}{c} \iint_{S_i} H_1^0 \cdot dS$$

$$= -\delta\mu_2 \cdot \overline{\overline{H}}_1^0 \tag{78}$$

It is obvious that from Eq. (43), $\delta j_2^{\alpha}$ should also be a closed current. Here, $\Delta I_i$ is a differential total current of $\delta j_2^{\alpha}$ which makes a fine closed loop of $C_i$ encircling an area $S_i$, and $\delta\mu_2$ is the magnetic moment of $\delta j_2^{\alpha}$.

From Eqs. (76) and (77), we have an identity of

$$\iiint_{V_2} \delta u_{kT}^{\alpha}\, dV + \iiint_{V_2} (A_2^0 + \frac{mc}{n_2 e^2} j_2^0) \cdot \frac{\delta j_2^{\alpha}}{c}\, dV + \iiint_{V_\alpha} [\frac{(\delta H^{\alpha})^2}{2} + \frac{m(\delta j_2^{\alpha})^2}{2n_2 e^2}]\, dV$$

$$+ \iiint_{V_\beta - V_\alpha} [\frac{(\delta E^{\alpha})^2}{2} + \frac{(\delta H^{\alpha})^2}{2}]\, dV = 0 \tag{79}$$

This means that conductor $C_2$ behaves just like as an independent subject. The sum of the variations of all the self energies $U_m$, $U_{kD}$ and $U_{e.m.}$ seems to become zero, irrespectively of the application of $H_1^0$ or $A_1^0$. We know in the previous equations from Eq. (63) to Eq. (79), the 1st order terms and the 2nd order terms should have separate equalities. We call the macroscopic transient electromagnetic fields of $\delta W_1^r$, MATE, because it represents a kind of macroscopic transient energy, whereas $\delta U_R^r$ will be called MITE, because it represents a microscopic transient energy. $\delta W_1^r$ could be positive or negative. When it is positive, then it can make a work to $C_1$, so that it has a capability to create an entropy of

$$\delta S = \frac{\delta W_1^r}{T} = \frac{[\text{MATE}]}{T}\,. \tag{80}$$

Here [MATE] = $\delta W_1^r$. On the other hand, when it is negative, an energy or a work has to be given outside from $C_1$, when MATE arrives at $C_1$. In this case MATE carries negative electromagnetic energy, or energy flows from outside to inside at the wave front $S_\beta$. $c\mathbf{E} \times \mathbf{H}$ directs towards P in Fig. 1, but yet the electromagnetic signal propagates towards outside, and no creation of entropy is expected.

For MITE, it is always positive and carries an entropy $\delta S_R$ to outside. Namely

$$[\text{MITE}] = \delta U_R^r = -\delta Q_R = -T\delta S_R \tag{81}$$

Further, let us denote the total transient energy $\delta U$ of Eq. (70) which was transfered from $C_2$ to the other systems as [TE]. Namely

$$[\text{TE}] = [\text{MATE}] + [\text{MITE}] = \delta W_1^r + \delta U_R^r. \tag{82}$$

From Eq. (75), we can say that MITE is located in $V_\beta - V_\alpha$. MATE, however, can not have such a simple interpretation, because, with the generation of $\delta \mathbf{j}_2$, the magnetic fields $\mathbf{H}_2^0$ which are located outside of $V_\beta$, become also not stable. Nevertheless, from Eq. (74), we may say that the action is located also in $V_\beta - V_\alpha$.

There is another typical initial situation at $t = t_0 + \delta t$, in which $V_\beta$ of Fig. 1 is located inside or nearly inside of $C_2$ and the diameter is $c\delta t$. Here $\delta t$ is assumed as the smallest time for the nucleus of $\delta \mathbf{j}_2$, or $\delta \mathbf{j}_N$, can be formed in a local part of $C_2$. It can be

$$\delta t = 1.7 \times 10^{-(15\sim 13)} \text{ sec} , \tag{83}$$

which is the time for an electron with the velocity $v = 10^{-(3\sim 5)}c$ can travel a distance of 5 Å. In this small time interval, the information of $\delta \mathbf{j}_N$ will travel a distance of

$$c\delta t \sim (0.5 \sim 50)\,\mu . \tag{84}$$

Then, at $S_\beta$ and from Eqs. (81) and (82), we have equations of

$$0 = -\iint_{S_\beta} c\delta\mathbf{E} \times \mathbf{H}^0 \cdot d\mathbf{S}\, dt = \iiint_{V_{\beta'}} [\mathbf{H}^0 \cdot \delta\mathbf{H} + \int_{t_0}^{t_0+\delta t} \delta\mathbf{E} \cdot \mathbf{j}^0\, dt]\ dV \tag{85}$$

and

$$0 = -\iint_{S_\beta} c\delta\mathbf{E} \times \delta\mathbf{H} \cdot d\mathbf{S}\, dt = \iiint_{V_\beta} [\frac{(\delta\mathbf{E})^2}{2} + \frac{(\delta\mathbf{H})^2}{2} + \int_{t_0}^{t_0+\delta t} \delta\mathbf{E} \cdot \delta\mathbf{j}_2 dt]\ dV \tag{86}$$

Therefore

$$\iiint_{V_\beta} \mathbf{H}^0 \cdot \delta\mathbf{H}\, dV = -\iiint_{V_\beta} \int_{t_0}^{t_0+\delta t} \delta\mathbf{E} \cdot \mathbf{j}^0\, dt\, dV = -(\delta U_{kT})_{(1)} - (\delta U_{kD})_{(1)} \tag{87}$$

and

$$\iiint_{V_\beta} \left[\frac{(\delta\mathbf{E})^2}{2} + \frac{(\delta\mathbf{H})^2}{2}\right] dV = -\iiint_{V_\beta} \int_{t_0}^{t_0+\delta t} \delta\mathbf{E} \cdot \delta\mathbf{j}_2\, dt\, dV$$

$$= -(\delta U_{kT})_{(2)} - (\delta U_{kD})_{(2)} > 0. \tag{88}$$

Here $(\;)_{(1)}$ and $(\;)_{(2)}$ indicate the 1st and 2nd order terms, respectively. In this situation,

$$\iiint_{V_\beta} \mathbf{H}^0 \cdot \delta\mathbf{H}\, dV = \iiint_{V_\beta} \mathbf{H}^0 \cdot \nabla \times \delta\mathbf{A}\, dV = \iiint_{V_\beta} \delta\mathbf{A} \cdot \frac{\mathbf{j}_2^0}{c} dV$$

$$= \iiint_{V_\beta} \mathbf{H}_2^0 \cdot \delta\mathbf{H}\, dV \tag{89}$$

Therefore

$$\iiint_{V_\beta} \mathbf{H}_1^0 \cdot \delta\mathbf{H}\, dV \equiv 0\ . \tag{90}$$

Now

$$c\nabla \times \delta\mathbf{H} = \delta\mathbf{j}_2 + \frac{\partial \delta\mathbf{E}}{\partial t} = \delta\mathbf{j}_2^* \tag{91}$$

and this indicates that the sum of the variation of the true currents and the displacement currents makes an effective closed current $\delta\mathbf{j}_2^*$, for which

$$\nabla \cdot \delta\mathbf{j}_2^* = 0\ . \tag{92}$$

It is to be noted however that, according to our assumption,

$$\frac{\partial \rho(\mathbf{r}, t)}{\partial t} = 0 \qquad \text{and} \qquad \nabla \cdot \mathbf{j}(\mathbf{r}, t) = 0 \tag{93}$$

even in a transient state. This indicates that

$$\nabla \cdot \delta \mathbf{E} = 0 \qquad \text{and} \qquad \nabla \cdot \delta \mathbf{j}_2 = 0 \tag{94}$$

in Eq. (91).

From Eq. (87) we have

$$\iiint_{V_\beta} \mathbf{H}^0 \cdot \delta \mathbf{H}\, dV = \iiint_{V_\beta} \mathbf{A}^0 \cdot \frac{\delta \mathbf{j}_2^*}{c}\, dV = \sum_i \frac{\Delta I_i^*}{c} \iint_{S_i} \nabla \times \mathbf{A}^0 \cdot d\mathbf{S} = \sum_i \delta \mu_i^* \cdot \overline{\overline{\mathbf{H}^0}}^{S_i}$$

$$= \delta \mu^* \cdot \overline{\overline{\mathbf{H}^0}} = \delta \mu^* \cdot \overline{\overline{\mathbf{H}_2^0}} \ , \tag{95}$$

as from Eq. (90)

$$\delta \mu^* \cdot \overline{\overline{\mathbf{H}_1^0}} = 0 \ . \tag{96}$$

Eqs. (95) and (96) show the presence of a very delicate situation. In Eq. (95) $\overline{\overline{\mathbf{H}_2^0}}$ can be even replaced by $\overline{\overline{\Delta \mathbf{H}_2^0}}$, which is the magnetic field due to the only closed loops of $\mathbf{j}_2^0$ which have a segment of the loops in $V_\beta$. Although $\delta \mathbf{j}_2$ must be controlled by the total magnetic field $\mathbf{H}^0 = \mathbf{H}_1^0 + \mathbf{H}_2^0$, the sign of the electromagnetic field energy created depends only on $\overline{\overline{\mathbf{H}_2^0}}$ of $\overline{\overline{\Delta \mathbf{H}_2^0}}$. When $\delta\mu^*$ is diamagnetic to $\overline{\overline{\mathbf{H}_2^0}}$ or $\overline{\overline{\Delta \mathbf{H}_2^0}}$, then from Eq. (87), $\delta U_k = \delta U_{kT} + \delta U_{kD}$ increases, whereas when $\delta\mu^*$ is paramagnetic to $\overline{\overline{\mathbf{H}_2^0}}$ or $\overline{\overline{\Delta \mathbf{H}_2^0}}$, then $\delta U_k$ decreases. This means that the increase in the magnetic moment due to $\mathbf{j}_2$ has to consume the thermal kinetic energy $U_{kT}$. This is the basic structure of the problem which has been already expected from Eq. (79).*

## § 7. Thermodynamical Derivation of the Meissner Effect of A Perfect Conductor.

In thermodynamics, we have two basic principles called the entropy maximum principle and the energy minimum principle.[29] The reference frame of the two principles is the thermodynamic configuration space in which the state of the system is represented by the possible macroscopic internal extensive parameters. Macroscopic dyanmical behavior is usually out of the problem in thermodynamics, so that the actual processes present cannot be obvious in thermodynamical analysis. The entropy maximum principle states that when the total energy $U^C$ has been fixed, the total entropy S of the equilibrium state is maximum. Namely, provided

*See p. 240–243 of Ref. 2. But thanks are due to Professor Takahashi, whose advice greatly enhanced the clear recognition of this fact by the author.

$$\delta U^C = 0 \ , \tag{97}$$

then in the equilibrium state

$$\delta S^C \leq 0 \ , \tag{98}$$

for the possible variation of the internal parameters in the configuration space. Here suffix C indicates that it is the quantity in the configuration space. On the other hand, the energy minimum principle states that provided

$$\delta S^C = 0 \tag{99}$$

then

$$\delta U^C > 0 \tag{100}$$

for the possible variation of the internal parameters in the configuration space.

It will be informed in advance that, using a physical conclusion that $\delta S^C = 0$, when $\delta U_{kT} = 0$ near the equilibrium state of our system comprising of $C_1$ and $C_2$, Eq. (100), applied for the local variation $\delta \mathbf{j}(\mathbf{r})$, is sufficient to derive the Meissner state. [34] However, in order to understand the implication of this result more deeply, we shall analyze the actual irreversible process in detail, thereby introducing a new thermodynamic principle.

Let us analyze the irreversible processes of a closed general system, which is enclosed strictly by a perfect reflector for the thermal radiation.

When the system considered is a macroscopic dynamic system and when initially the system is constrained to be at rest in a macroscopically non-stable configuration, then, due to the general basic dyanmical principle in physics, the system starts variation towards the direction in which $U^C$ decreases, in agreement with Eq. (100). At the same time, the surplus energy

$$-\delta U^C > 0 \tag{101}$$

becomes in a form of transient energy, TE, such as macroscopic kinetic energy, electromagnetic rediation energy, etc. A part of TE can be a very localized heat also. Keeping this concept of

$$[\mathrm{TE}] = -\delta U^C \tag{102}$$

in mind, let us convert the implications of the two principles in terms of irreversible processes. The entropy maximum principle requests that, in a process, when

$$\delta U^C = 0 \qquad \text{or} \qquad [\mathrm{TE}] = 0 \tag{103}$$

then

$$\delta S^C > 0 \tag{104}$$

is a "necessary" condition that the assumed process is an irreversible process. Similarly, the energy minimum principle requests that, in a process, when

$$\delta S^{C} = 0 \tag{105}$$

then

$$\delta U^{C} < 0 \qquad \text{or} \qquad [TE] = -\delta U^{C} > 0 \tag{106}$$

is a "necessary" condition that the assumed process is irreversible. For real processes, we can only say about the "necessary" condition, because we don't know the details of the mechanism. This condition, however, is usually sufficient to get the conclusion, because in a real problem we know the mechanism. Although Eq. (101) is derived from macroscopic dynamics and Eq. (106) from microscopic thermodynamics, (29) two results are essentially identical, showing that the principle is quite general and participation of persistent currents does not violate this principle, as in the case of several multiply connected superconductors. 30)

Our essential problem now is to find out a more general principle in which dynamical principle and thermal principle may compete. We claim that

$$[TE] + T\delta S > 0 \tag{107}$$

is a "necessary" condition that the process is irreversible. We call this thermodynamic principle as the transient energy principle. The proof is as follows. In a strictly closed system and in a simple system in which macroscopic dynamic principle operates

$$[TE] = -\delta U^{C} \geq 0 \ . \tag{108}$$

The possible additional entropy, $\delta S^*$, that the system will get when virtually all the reactions of [TE] have finished, must be

$$\delta S^* \leq \frac{[TE]}{T} \ , \qquad \delta U^{C*} = 0 \tag{109}$$

Therefore, when the process is irreversible, we must expect from Eq. (98) that

$$0 < \delta S + \delta S^* \leq \delta S + \frac{[TE]}{T} \tag{110}$$

Namely, Eq. (107) is a "necessary" condition that the process is irreversible in this simple system.

As is seen easily, this principle includes the mentioned two principles of Eqs. (103) – (106). Further, when we could assume that the system is in contact with a heat reservor at $T^{r}$

and is near the equilibrium state, the Helmholtz free energy principle states that

$$\delta U^{C} - T^{r}\delta S < 0 \tag{111}$$

is a "necessary" condition that the process is irreversible.[33] Eg. (111) is identical to Eq. (107), except the difference that whether the surplus energy is absorbed in the heat reservor or not. Since the equilibrium state of a system itself should be defined independently of the presence or absence of the heat reservor, when we regard Eqs. (107) and (111) as the necessary relations in the configuration space for the irreversible process, they are essentially identical and, relying on the analytical structure of the physical system, [31], [32] we conclude that Eq. (107) is a "necessary" condition in general that the process is irreversible. In Eq. (107), a very localized heat can be either in [TE] or in T$\delta$S, which does not affect the principle, indicating that the principle satisfies this necessary requirement.

Our system presents a delicate example of this principle. The dynamic principle which is operating in our system is the diamagnetic cyclotron motion of each electron in a magnetic field. With Eq. (107), mathematically, we have expanded the "necessary" condition into the region where $\delta S < 0$ or $[TE] < 0$. Whether these ultimate situations can happen or not is a different problem, because Eq. (107) is only a "necessary" condition and it depends on whether the initial state assumed is realizable or not. We mention, however, in our case, when the system approaches thermal equilibrium, the [TE] for the assumed elementary unit process converges to a thermal radiation, so that $\delta S$ in this limitting situation has to become negative. It is also mentioned that, when we could have a very thin completely diamagnetic surface current state initially, then the following change has to become $[TE] = -\delta U^{C} < 0$.

It will be mentioned that in a simple thermodynamic system in which there is only one extensive parameter, X,[29] Eq. (107) implies that the change of X in the configuration space is always towards the equilibrium value $X^{e}$.

One interesting application of this principle is a compressed gas in a cylinder with a piston supported by a spring. In an idealized situation,

$$U = U_{apring} + U_{gas} \tag{112}$$

$$\delta S = \delta S_{gas} = 0 \tag{113}$$

Therefore

$$[TE] = -\ [\ \delta U^{C}_{spring} + \delta U^{C}_{gas}\ ] > 0 \tag{114}$$

is the condition that the process is irreversible. It is noted that $U_{kT}$ can convert to a work in this system, which, as will be shown soon, is quite similar to our case.

In order to apply TE principle to our problem, let us concentrate our attention to a local volume of a large system. Our main interest is the thermally activated and/or dynamically driven tiny irreversible macroscopic change of localized internal extensive parameters. TE in this case is the transient energy created as a result of such an extremely small irreversible macroscopic process. Then we have

$$\begin{aligned}[TE] + T\delta S &= -\delta U^{C} + T\delta S_{V} \\ &= -\ \delta(U_{V} - T^{r}S_{V}) > 0\end{aligned} \tag{115}$$

We have defined the volume V as the smallest volume which includes all the regions in which the change in the internal extensive parameters have happened. $\delta U^{C} = \delta U_{V}$ must include the change due to the long range interactions. Eq. (115) is identical to the minimum Helmholtz free energy requirement in an isothermal process. This must be, since the local volume, V, can be regarded as a system in a heat reservor with the temperature $T^{r}$. Here we have carefully differentiated Eq. (115) from the other thermodynamic relation of the Helmholtz free energy, i.e.,

$$\delta F_{V} = \delta A_{V} + \delta Q_{V} - T\delta S_{V} = \delta A_{V} < 0 \tag{116}$$

because the elementary process that we are considering is not quasi-static and the work done to the outside, $(-\delta A_{V})$, is not always well defined at least at the instant when the process has just finished. The transient energy principle of Eq. (115) is a slight logical extention of Eq. (116).

Now let us discuss our problem. In this case,

$$[TE] = [MATE] + [MATE] \tag{117}$$

can be defined quite clearly. But $T\delta S$ is not known analytically.

Physically, when

A), $\quad \delta U_{kT} < 0,$ $\tag{118}$

then

$$\delta S \leq 0 \tag{119}$$

and

$$|\delta U_{kT}| \geq |T\delta S| \quad \text{or} \quad \delta U_{kT} \leq T\delta S \tag{120}$$

Namely in this case

$$[TE] + T\delta S \geq [TE] + \delta U_{kT} \tag{121}$$

Therefore

$$[TE] + \delta U_{kT} > 0 \tag{122}$$

is a sufficient condition that the necessary condition is satisfied. When, however,

B), $$\delta U_{kT} > 0, \tag{123}$$

then

$$\delta S \geq 0 \tag{124}$$

so that

$$|\delta U_{kT}| \geq |T\delta S| \quad \text{or} \quad \delta U_{kT} \geq T\delta S \tag{125}$$

Therefore in this case we have

$$[TE] + \delta U_{kT} \geq [TE] + T\delta S > 0 \tag{126}$$

There is an important physical requirement in our system. When the system approaches to an equilibrium state and, if we make the magnitude of the variation $\delta j_N$ small, then the process must converge to a purely thermal heat radiation from the system. This means that in these limitting situations,

$$\delta U_{kT} \rightarrow T\delta S = -\delta Q_R < 0, \quad [TE] = \delta Q_R \tag{127}$$

Here, $\delta Q_R$ is the heat radiated. Therefore, when approaching to the equilibrium, $\delta U_{kT}$ approaches to $T\delta S$ rapidly and we have the relation

$$[TE] + T\delta S = [TE] + \delta U_{kT} = \delta Q_R - \delta Q_R = 0 \tag{128}$$

This relation is sufficient to conclude the Meissner state as shown later. When the system is not near the equilibrium and the magnetic field **H** has penetrated into system $C_2$ sufficiently, then when $\delta j_N$ is diamagnetic, as will be shown soon, we have case A) and

$$[TE] + T\delta S \geq [TE] + \delta U_{kT} > 0 \tag{129}$$

On the other hand, when $\delta j_N$ is paramagnetic, we have case B) and

$$0 > [TE] + \delta U_{kT} \geq [TE] + T\delta S \tag{130}$$

Therefore, our necessary condition of Eq. (107) is sufficient to conclude that the irreversible process must be towards the Meissner state.

Let us show the result analytically. The energy identity at $t = t_0 + \delta t$ must be

$$\delta U^C_m + [\mathrm{MATE}] + \delta U^C_{kD} + \delta U_{kT} + [\mathrm{MITE}] = 0 \tag{131}$$

$$\left.\begin{aligned}
&\delta U^C_m = \iiint_{V_2} \mathbf{A}^0 \cdot \frac{\delta j_2^\alpha}{c}\, dV + \iiint_\infty \frac{(\delta \mathbf{H})^2}{2}\, dV \\
&[\mathrm{MATE}] = \delta W^r_1, \qquad [\mathrm{MITE}] = \delta U^r_1 \\
&\delta U^C_{kD} + \delta U_{kT} = \int_{t_0}^{t_0+\delta t} \iiint_{V_2} \delta \mathbf{E} \cdot j_2\, dV dt
\end{aligned}\right\} \tag{132}$$

Then we have

$$[\mathrm{TE}] + \delta U_{kT} =$$

$$[\mathrm{MATE}] + [\mathrm{MITE}] + \delta U_{kT} = -(\delta U^C_m + \delta U^C_{kD})$$

$$= -\iiint_{V_\beta} (\mathbf{A}^0 + \frac{mc}{n_2 e^2} j_2^0) \cdot \frac{\delta j_N}{c} dV - \iiint_{V_\beta} \frac{\delta \mathbf{A}_N \cdot \delta j_N}{2c} dV - \iiint_{V_\beta} \frac{m}{n_2 e^2} \frac{(\delta j_N)^2}{2} dV \tag{133}$$

We know that the last two integrals of Eq. (133) cancel out mutually. Namely, neglecting higher order terms, we get from Eq. (62) in general

$$-\frac{d}{dt}\frac{m}{ne^2}\delta j = -\mathbf{E} + \frac{j}{ce} \times \mathbf{H} = \nabla \varphi + \frac{1}{c}\frac{\partial \mathbf{A}}{\partial t} + \frac{j}{ce} \times \mathbf{H} \tag{134}$$

Let us separate the vector potential $\mathbf{A}$ into

$$\mathbf{A} = \mathbf{A}^I + \nabla \psi^I , \tag{135}$$

in which $\mathbf{A}^I$ has no surface normal component at the surface of the conductor. $\mathbf{A}^I$ will be called Iida's gauge vector potential hereafter. Separating the current $\delta j$ into a very small polarization current component $\delta j^P$ and a persistent current component $\delta j^C$, we get

$$-\frac{m}{ne^2}\frac{d\delta j^P}{dt} = \nabla (\varphi + \frac{1}{c}\frac{\partial \psi^I}{\partial t}) + \frac{j}{ec} \times (\nabla \times \mathbf{A}^I) \tag{136}$$

$$-\frac{m}{ne^2}\frac{d\delta j^C}{dt} = \frac{1}{c}\frac{\partial \delta A^I}{\partial t} \tag{137}$$

$$\delta A^I = -\frac{mc}{ne^2}\delta j^C \tag{138}$$

putting $\delta j^C$ and $\delta A$ as $\delta j_N$ and $\delta A_N$, we can get an exact cancellation in Eq. (133). *

Now, at the equilibrium, from Eq. (128), we must expect that Eq. (133) is zero. Then the first integral can be converted as

$$-\iiint_{V_\beta} (A^0 \pm \frac{mc}{n_2 e^2} j_2^0) \cdot \frac{\delta j_N}{c}\, dV$$

$$= -\sum_i \frac{\Delta I_i}{c} \iint_{S_i} \nabla \times (A^0 + \frac{mc}{n_2 e^2} j_2^0) \cdot dS = 0 \quad . \tag{139}$$

When we put

$$\nabla \times (A^0 + \frac{mc}{n_2 e^2} j_2^0) = \mathbf{H}^* \ , \tag{140}$$

then

$$-\iiint_{V_\beta} (A^0 + \frac{mc}{n_2 e^2} j_2^0) \cdot \frac{\delta j_N}{c}\, dV = -\delta\mu_N \cdot \overline{\overline{\mathbf{H}}}^* = 0 \tag{141}$$

Then, the necessary condition for the equilibrium is

$$\mathbf{H}^* = \nabla \times (A^0 + \frac{mc}{n_2 e^2} j_2^0) = 0 \tag{142}$$

so that

$$A^0 + \frac{mc}{n_2 e^2} j_2^0 = \nabla \psi \tag{143}$$

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*From Eq. (138), we get

$$\Box^2 \delta A^I = -\frac{\delta j^C}{c} = \frac{ne^2}{mc^2}\, \delta A^I \quad .$$

Applying a Fourier transformation, we can get the penetration depth of $\delta j^C$ as in the order of $\Lambda = mc^2/ne^2$.

in which $\psi$ (r) is an arbitrary scalar function in $V_2$. Eq. (143) is known as a London condition and we know that Eq. (143) is sufficient to conclude the Meissner state uniquely in $C_2$. We believe that Eqs. (141) and (142) are essentially identical to the equation derived by de Gennes.[34]

It will be obvious from Eq. (133), that we have Eqs. (129) and (130) for the process when $\overline{\overline{\mathbf{H}}}^*$ is along **H**.

## § 8. Kinematical Stability and the Energy Relation of the Meissner State

Although the derivation of the Meissner state in § 7 stands on purely thermodynamic calculation, we know that the state is kinematically stable.[26] The time change of $\mathbf{p}_i$ of each electron is described by

$$\frac{d\mathbf{p}_i}{dt} = \nabla_i \left[\frac{q_i\mathbf{v}_i}{c}\cdot \mathbf{A}_1(\mathbf{r}_i)\right] - q_i\nabla_i\varphi_1(\mathbf{r}_i) \ . \tag{144}$$

Here, we are watching a single electron which is intenerating randomly in $C_1$, and we have neglected the time dependent Maxwell–Lorentz fluctuating part of the electromagnetic potentials. With using London gauge

$$\mathbf{A}_1^L(\mathbf{r}_i) = \mathbf{A}_1(\mathbf{r}_i) - \nabla_i\psi_1(\mathbf{r}_i) \tag{145}$$

we get

$$\frac{d\mathbf{p}_i^L}{dt} = \nabla_i \left[\frac{q_i\mathbf{v}_i}{c}\cdot \mathbf{A}_1{}^L(\mathbf{r}_i)\right] - q_i\nabla_i\varphi_1'(\mathbf{r}_i) \ , \tag{146}$$

so that

$$\frac{d\overline{\mathbf{p}}_i^L}{dt} = \nabla_i \left[\frac{q_i\overline{\mathbf{v}}_i}{c}\cdot \mathbf{A}_1{}^L(\mathbf{r}_i)\right] - q_i\nabla_i\varphi_1(\mathbf{r}_i) \tag{147}$$

Here, single bar means time average. Now, since $\mathbf{A}_1{}^L(\mathbf{r}_i)$ and $\overline{\mathbf{v}}_i$ in the Meissner state are parallel to the surface and the main term of $\varphi_1(\mathbf{r}_i)$ is due to the presence of the surface boundary, Eq. (147) predicts that there is no change in the surface parallel component of $\overline{\mathbf{p}}_i^L$ Then

$$[\overline{\mathbf{p}}_i^L]_{/\!/} = [m_i\overline{\mathbf{v}}_i + \frac{q_i}{c}\mathbf{A}_1{}^L(\mathbf{r}_i)]_{/\!/} \tag{148}$$

is a constant of motion. When electron i comes from the region where $\bar{v}_i$ and $A_I^L = 0$, then Eq. (148) tells

$$[m_i \bar{v}_i]_{/\!/} = [-\frac{q_i}{c} A_I{}^L(r_i)]_{/\!/} \tag{149}$$

From Eq. (149), Eq. (147) predicts that the surface normal components of the velocities are identical with different signs for the incoming and outgoing electrons. This means that the volume average of the surface normal component of the velocity is zero. Therefore, Eq. (149) is identical to the London equation.

From Eq. (48), the magnetic energy $U_{1m}$ of the system $C_1$ is

$$U_{1m} = \iiint_{\infty} \frac{H_1^2}{2} dV = \iiint_{V_1} \frac{1}{2c} \mathbf{j}_1 \cdot \mathbf{A}_1^L \, dV + \iint_{S_1^*} \frac{\mathbf{j}_1}{2c} \psi_1(\mathbf{r}) \cdot d\mathbf{S} \ .$$

$$= \iiint_{V_1} \frac{1}{2c} \mathbf{j}_1 \cdot \mathbf{A}_1^L \, dV + \frac{1}{2c} I_1 \Phi_1 \ . \tag{150}$$

Here $S_1{}^*$ is the surface of a simply connected body $V_1{}^*$ obtained from $V_1$ by removing a thin cross sectional plate. Since, we have an identity

$$n_1 \frac{mv_D^2(\mathbf{r})}{2} + \frac{\mathbf{j}_1(\mathbf{r}) \cdot \mathbf{A}_1{}^L(\mathbf{r})}{2c} = 0 \ , \tag{151}$$

we have

$$U_1 = U_{1T} + \iiint_{V_1} \frac{m}{2n_1 e^2} j_1^2 \, dV + U_{1m} = U_{1T} + \frac{1}{2c} I_1 \Phi_1 \ . \tag{152}$$

Here $U_{1T}$ is the thermal part of the energy. The relation can be extended easily to a general system as

$$U_{total} = \sum_{\ell} U_{\ell T} + \sum_{\substack{\ell \\ \beta}} \frac{1}{2c} I_{\beta^\ell} \Phi_{\beta^\ell} \ . \tag{153}$$

in which, $\beta^\ell$ is the number of a hole in $V_\ell$, $\Phi_{\beta^\ell}$ is the total magnetic flux confined in it, and $I_{\beta^\ell}$ is the total current which are circling arround the hole.[35), 36), 37), 38)]

## § 9. Thermodynamic Functions of the Meissner State

Let us convert a part of our notations from **A(r)** and **j(r)** to **H(r)** and calculate the energy $U_V$ of a small volume V in $C_2$ in terms of the Maxwell–Lorentz fields. Then

$$U_V^{II} = \iiint_V \frac{e^2 + h^2}{2} dV = \sum_\lambda \iiint_V \frac{e_\lambda^2 + h_\lambda^2}{2} dV + \sum_i \iiint_V \frac{e_i^2 + h_i^2}{2} dV$$

$$+ \sum_{\lambda \neq \mu} \iiint_V \frac{\mathbf{e}_\lambda \cdot \mathbf{e}_\mu}{2} dV + \sum_{i \neq j} \iiint_V \frac{\mathbf{e}_i \cdot \mathbf{e}_j}{2} dV + \sum_{\lambda, i} \iiint_V \mathbf{e}_\lambda \cdot \mathbf{e}_i \, dV$$

$$+ \sum_{\lambda \neq \mu} \iiint_V \frac{\mathbf{h}_\lambda \cdot \mathbf{h}_\mu}{2} dV + \sum_{i \neq j} \iiint_V \frac{\mathbf{h}_i \cdot \mathbf{h}_j}{2} dV + \sum_{\lambda, i} \iiint_V \mathbf{h}_\lambda \cdot \mathbf{h}_i \, dV \, . \qquad (154)$$

Here the charges in V are designated by i and all the other charges are by λ. Let us assume that V is a very small thin shell volume in $C_2$ in which all physical quantities can be assumed uniform. Then, similarly to Eq. (7) and assuming a unit volume to V for convenience we get

$$u^{II} = \sum_i m_i c^2 + \sum_i \frac{m_i}{2} v_i^2 + \sum_i \frac{3 m_i v_i^4}{8c^2} + \frac{\rho(\mathbf{r})\,\varphi(\mathbf{r})}{2} + U_{E.S.f.}$$

$$+ \frac{H^2}{2} + U_{M.f.} - \frac{1}{2c^2}\left(\frac{\partial \varphi}{\partial t}\right)^2 + U_{E.D.f.} \qquad (155)$$

$$= u^I + \frac{H^2}{2} \qquad (156)$$

Regarding

$$\frac{\rho(\mathbf{r})\,\varphi(\mathbf{r})}{2}, \; U_{E.S.f.}, \; U_{M.f.}, \; -\frac{1}{2c^2}\left(\frac{\partial \varphi}{\partial t}\right)^2, \; U_{E.D.f.}$$

and the first and second terms of Eq. (48) as a part of the lattice energy $U_{L2}$ and the kinetic energy $\sum_i U_{k2}^i$ of Ref. 37) we get

$$u^{II} = u_L + \sum_i u_{k2}^i + \frac{H^2}{2} \qquad (157)$$

which is identical to the total internal energy of the system of Eq. (78) in Ref. 37). In Eq. (156), we show the mutual relation between $u^I$ and $u^{II}$.

In Eq. (55), if we consider a quasi-static change of the equilibrium state

$$\iiint_V \int_{t_0}^{t_0+\delta t} \mathbf{E}(\mathbf{r}, t) \cdot \mathbf{j}(\mathbf{r}, t)\, dt\, dV = \iiint_V \int_{t_0}^{t_0+\delta t} \left(-\frac{1}{c}\frac{\partial \mathbf{A}}{\partial t} - \nabla \varphi\right) \cdot \mathbf{j}\, dt\, dV$$

$$= \iiint_V \left[ -\frac{1}{c}\delta \mathbf{A} - \nabla \int_{t_0}^{t_0+\delta t} \varphi(\mathbf{r}, t)\, dt \right] \cdot \mathbf{j}\, dV \qquad (158)$$

Applying Iida's gauge, $\mathbf{A}(\mathbf{r}, t)$ in $C_2$ (when simply connected) can be separated uniquely into

$$\mathbf{A}(\mathbf{r}, t) = \mathbf{A}^I(\mathbf{r}, t) + \nabla \psi^I(\mathbf{r}, t) \ . \qquad (159)$$

Then, when $\mathbf{A}(\mathbf{r}, t)$ is changing slowly with time, the component of the induced electric field, which can not circulate in $C_2$, should be instantly compensated by the induced depolarization electric field $-\nabla \varphi(\mathbf{r}, t)$, which has been generated by the instant extremely small shift of all the electrons. Namely the total effect is

$$-\frac{1}{c}\frac{\partial \mathbf{A}}{\partial t} - \nabla \varphi = -\frac{1}{c}\frac{\partial \mathbf{A}^I}{\partial t} - \frac{1}{c}\frac{\partial \nabla \psi^I}{\partial t} - \nabla \varphi = -\frac{1}{c}\frac{\partial \mathbf{A}^I}{\partial t} \ . \qquad (160)$$

$$\varphi(\mathbf{r}, t) = -\frac{1}{c}\frac{\partial \psi^I(\mathbf{r}, t)}{\partial t} \ . \qquad (161)$$

Therefore we get in Eq. (158)

$$\delta \mathbf{A} + c\nabla \int_{t_0}^{t_0+\delta t} \varphi(\mathbf{r}, t)\, dt = \delta \mathbf{A} - \nabla \int_{t_0}^{t_0+\delta t} \frac{\partial \psi^I(\mathbf{r}, t)}{\partial t} dt$$

$$= \delta \mathbf{A} - \nabla \delta \psi^I(\mathbf{r}, t) = \delta \mathbf{A}^I \qquad (162)$$

Since the above-mentioned shift has no appreciable work, we get from Eqs. (157) and (158),

$$du^{II} = Tds + \mathbf{H} \cdot d\mathbf{H} - \frac{\mathbf{j}}{c} \cdot d\mathbf{A}^I \ . \qquad (163)$$

Here, we have assumed the heat input (or output) dQ as

$$dQ = Tds. \qquad (164)$$

This means that we have put $\delta P_T = 0$ in Eq. (64) for this quasi-static reversible change. Further

$$d(u^{II} - Ts + \frac{\mathbf{j}}{c} \cdot \mathbf{A}^I) = -sdT + \mathbf{H} \cdot d\mathbf{H} + \mathbf{A}^I \cdot \frac{d\mathbf{j}}{c} . \tag{165}$$

Then from Eq. (165), after a certain calculation, we can conclude that a kind of the Gibbs function

$$g^{II} = u^{II} - Ts + \frac{\mathbf{j}}{c} \cdot \mathbf{A}^I = u_L + \sum_i u^i_{k2} + \frac{H^2}{2} + \frac{\mathbf{j}}{c} \cdot \mathbf{A}^I - Ts \ . \tag{166}$$

is a constant over the entire volume of $C_2$. The logics is as follows. At a fixed location, when the Meissner state has been realized, then

$$\mathbf{A}^I \rightarrow \mathbf{A}^L = -\frac{\Lambda^2}{c} \mathbf{j} \tag{167}$$

$$\mathbf{H} = \nabla \times \mathbf{A}^I = -\frac{\Lambda^2}{c} \nabla \times \mathbf{j} \tag{168}$$

$$\mathbf{j} = \mathbf{j}_0 \exp\left[-\frac{\zeta}{\Lambda}\right] , \tag{169}$$

Therefore, in general,

$$\mathbf{H} \cdot d\mathbf{H} + \mathbf{A}^L \cdot \frac{d\mathbf{j}}{c} = d\left[\frac{\Lambda^4}{c^2} \frac{(\nabla \times \mathbf{j})^2}{2} - \frac{\Lambda^2}{c^2} \frac{\mathbf{j}^2}{2}\right] = 0 , \tag{170}$$

Then we get

$$dg^{II} = d(u^{II} - Ts + \frac{\mathbf{j}}{c} \cdot \mathbf{A}^L) = -sdT \ . \tag{171}$$

Thus, when the temperature is kept constant, we can assume that

$$\begin{aligned} g^{II} = u^{II} - Ts + \frac{\mathbf{j}}{c} \cdot \mathbf{A}^L &= u_L + \sum_i u^i_{k2} + \frac{1}{2}\frac{\mathbf{j}}{c} \cdot \mathbf{A}^L - Ts \\ &= u_L + \sum_i u^i_{k2} - \frac{H^2}{2} - Ts \end{aligned} \tag{172}$$

is a constant over the entire volume of $C_2$.

This is the rigorous derivation of the relation of Eq. (83) of our paper part II[2], where the derivation was in an exploratory stage and was not completely correct. From Eq. (172), we get

$$\Delta(\sum_i u^i_{k2}) - T\Delta s = \frac{H^2}{2} = -\frac{\mathbf{j}\cdot\mathbf{A}^L}{2c} \tag{173}$$

where $\Delta$ means the increase from the value in the interior region where $\mathbf{H} = 0$. Since from Eq. (151)

$$-\frac{\mathbf{j}\cdot\mathbf{A}^L}{2c} = n_2 \frac{m}{2} v_D^2 = \Delta u_{kD}, \tag{174}$$

we get

$$[\,\Delta(\Sigma u^i_{k2}) - \Delta u_{kD}\,] - T\Delta s = 0, \qquad \frac{\Delta s}{\Delta[\,\Sigma u^i_{k2} - u_{kD}\,]} = \frac{1}{T} \tag{175}$$

Eq. (175) is a reasonable expression. Since the direct action of the magnetic field does not change $\Sigma u^i_{k2}$, the subtraction of $\Delta u'_{kD}$ will probably result in slightly negative $\Delta[\Sigma u^i_{k2} - u_{kD}]$ and $\Delta s$. Nevertheless we can conclude that the Gibbs-Duhem relation on the phase equilibrium seems valid inside of a perfect conductor and the unit volume Gibbs function $g^{II}$ is a constant over the entire volume of $C_2$, irrespectively of the presence or absence of $\mathbf{H}(\mathbf{r})$. At the critical magnetic field $H_c(T)$ for the super to normal transition, w should expect

$$g^n(H_c) = g^s(H_c) = g^s(0)\ . \tag{176}$$

Here n and s indicate normal and super respectively. Therefore from Eqs. (166) and (171),

$$f^n(H_c) - H_c^2 = f^n(0) - \frac{H_c^2}{2} = f^s(0) \tag{177}$$

in which f means the unit volume Helmholtz free energy and we have assumed

$$f^n(H) = f^n(0) + \frac{H^2}{2}\ , \qquad g^n(H) = f^n(H) - H^2 \quad (B \fallingdotseq H) \tag{178}$$

according to Eqs. (71) and (73) of the thermodynamics of a diamagnet presented in Ref. 2).

From the argument that we have developed, the total thermodynamic function to be minimized at constant temperature and applied magnetic field will be the Helmholtz free energy which can be represented as

$$F^I = U_2^I - TS_2 \qquad (\text{ or } F^{II} = U_2^{II} - TS_2) \tag{179}$$

in which $U_2^I$ is given by

$$U_2^I = \sum_i m_i c^2 + \sum_i \frac{m_i}{2} v_i^2 + \sum_{i>j} \frac{q_i q_j}{4\pi r_{ij}} + \sum_{i>j} \frac{q_i q_j (\mathbf{v}_i \cdot \mathbf{v}_j)}{4\pi r_{ij} c^2} + \sum_{i,\lambda} \frac{q_i q_\lambda}{4\pi r_{i\lambda}} + \sum_{i,\lambda} \frac{q_i q_\lambda \mathbf{v}_i \cdot \mathbf{v}_\lambda}{4\pi r_{i\lambda} c^2}$$

$$= \text{const} + \iiint_{V_2} \frac{n_2 m}{2} \overline{(v_i - v_D)^2}\, dV + \iiint_{V_2} \left[\frac{m}{2n_2 e^2} j_2^2 + \frac{\mathbf{j}_2 \cdot \mathbf{A}_2}{2c} + \frac{\mathbf{j}_2 \cdot \mathbf{A}_1}{c}\right] dV \tag{180}$$

Let us confirm this. Namely we have

$$\delta F^I = \left(\frac{\partial U_2^I}{\partial S_2}\right)_{A_1(\mathbf{r}), j_2(\mathbf{r})} \delta S_2 + (\delta U_2^I)_{S_2} - S_2 \delta T - T\delta S_2$$

$$= (\delta U_2^I)_{S_2} - S_2 \delta T \tag{181}$$

We know from Eqs. (133) and (139), in general in the configuration space

$$(\delta U_2^I)_{S_2} = (\delta U_2^I)_{S_2, \mathbf{A}_1(\mathbf{r})} + (\delta U_2^I)_{S_2, j_2(\mathbf{r})}\ , \tag{182}$$

but from the condition of the equilibrium,

$$(\delta U_2^I)_{S_2, \mathbf{A}_1} = 0 \tag{183}$$

for the equilibrium states. Therefore we get

$$\delta F^I = (\delta U_2^I)_{S_2, j_2(\mathbf{r})} - S_2 \delta T \tag{184}$$

which indicates that $F^I$ should be minimized at constant temperature and applied field. With using our approximation, the explicit forms of these equations are,

$$(\delta U_2^I)_{S_2, \mathbf{A}_1} = \iiint_{V_2} \left[\frac{m}{n_2 e^2} \mathbf{j}_2 + \frac{\mathbf{A}_2 + \mathbf{A}_1}{c}\right] \cdot \delta \mathbf{j}_2\, dV = 0, \tag{185}$$

$$(\delta U_2^I)_{S_2, j_2(\mathbf{r})} = \iiint_{V_2} \frac{\mathbf{j}_2}{c} \cdot \delta \mathbf{A}_1\, dV\ . \tag{186}$$

Therefore

$$\delta F^I = \iiint_{V_2} \frac{j_2}{c} \cdot \delta A_1 \, dV - S_2 \delta T \quad . \tag{187}$$

This confirms our previous conclusion of Eq. (104) of Ref. 2).

§ 10. Conclusions and other Discussions

Now we have established a concept that a moving electron associates scalar and vector potentials which are combined to the electron pseudo-elastically. The presence of an energy term $-e\mathbf{V}\cdot\mathbf{A}(\mathbf{r})/c$ is established in thermodynamics independently of the conventional Hamiltonian of the electron. As has been shown in the studies of the foregoing two papers, this introduction of a new energy term in the physics of magnetism will result in many new features, not only in solid state physics, but could be also in the other fields. Extending the energy minimum principle in thermodynamics, a new thermodynamic principle, called the transient energy principle has been established. From this principle, it has been proved that the Meissner effect should be a classical property of perfect conductors, so that Miss Van Leeuwen's theorem must be wrong. [1), 2)] New thermodynamics[2)] should be used for the magnetizable materials and superconductors. Thermostatistics of magnetizable materials must differentiate the Larmor diamagnet and the Curie-Langevin-Debye paramagnet, since the transfer relations of the electromagnetic energy in these two materials are essentially different[2)]. Although the old understanding was very poor[39)], there is a fortunate situation that the old logics is still approximately correct in cases when the diamagnetic susceptibility is quite small[2)] (e.g., the Landau diamagnetism). As shown in Eqs. (5), (7), and (18), when a collective electrons is confined in a certain volume, as is the case of a superconductor, the Coulomb term will be already in its minimum state and a new term

$$\sum_{i>j} \frac{q_i q_j}{4\pi r_{ij}} \frac{\mathbf{v}_i \cdot \mathbf{v}_j}{c^2} + \sum_{i>j} \frac{q_i q_j}{8\pi r_{ij}} \frac{\mathbf{v}_i \times \hat{\mathbf{r}}_{ij} \cdot \mathbf{v}_j \times \hat{\mathbf{r}}_{ji}}{c^2}$$

$$= \sum_{i>j} \frac{q_i q_j}{8\pi r_{ij}} \frac{[\, \mathbf{v}_i \cdot \mathbf{v}_j - v_i v_j \, \mathrm{cas}\psi_{ij} \mathrm{cas}\psi_{ji} \,]}{c^2} \tag{188}$$

may become quite important. Here $\psi_{ij}$ is the angle between $\mathbf{r}_{ji}$ and $\mathbf{v}_i$. This term becomes minimum when each two electrons forms a pair having opposite velocities and run on an identical straight line. This term may play a certain important role for the creation of the Cooper pair in superconductors. The magnitude becomes

$$\frac{e^2}{4\pi r}\left(\frac{v}{c}\right)^2 \sim 7°K \tag{189}$$

for the separation of 2Å and

$$\frac{v}{c} \sim 10^{-2} \ . \tag{190}$$

Quantum mechanical version of our analyses must be developed.[40] Although our description in this paper does not use quantum theoretical argument, the essential physics clarified should not depend on the way of the description, either classically or quantally. Therefore, the old quantum description of the Meissner state [41] must be not entirely correct, or has disregarded a certain group of electronic states. The electronic states of the paramagnetic boundary electrons, which had an essential rôle in Miss Van Leeuwen's theorem, will be such electronic states, the energies of which become very high in our theory but should not be so high in the old treatment. We suggest in this respect that a study on the magnetism of superconducting fine particles will present a good justification of our theory.

Applications of these results for the plasma physics and astronomical physics are also proposed. There might be a possibility that the new term has an important role for the creation of the earth magnetic field either in the interior or in the outside ion atmosphere of the earth.

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